1123

PB 210 018

Final Report

REGIONAL AIR POLLUTION STUDY: A PROSPECTUS

Part II — Research Plan

Prepared for:

THE ENVIRONMENTAL PROTECTION AGENCY
NATIONAL ENVIRONMENTAL RESEARCH CENTER
RESEARCH TRIANGLE PARK, NORTH CAROLINA

CONTRACT 68-02-0207

SRI Project 1365

STANFORD RESEARCH INSTITUTE
Menlo Park, California 94025 · U.S.A.

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
Springfield, Va. 22151

280

BIBLIOGRAPHIC DATA SHEET

1. Report No. APTD-1123

2.

3. Recipient's Accession No.

4. Title and Subtitle

Regional Air Pollution Study: A Prospectus
Part II - Research Plan

5. Report Date
January 1972

6.

7. Author(s)

The report describes the research plan to be used in the (RAP) which is being developed to aid in improving and controlling regional air quality.

8. Performing Organization Rept. No. SRI Project 1365

9. Performing Organization Name and Address

Stanford Research Institute
Menlo Park, California 94025

10. Project/Task/Work Unit No.

11. Contract/Grant No.
68-02-0207

12. Sponsoring Organization Name and Address

THE ENVIRONMENTAL PROTECTION AGENCY
NATIONAL ENVIRONMENTAL RESEARCH CENTER
RESEARCH TRIANGLE PARK, NORTH CAROLINA

13. Type of Report & Period Covered
Final

14.

15. Supplementary Notes

16. Abstracts

A comprehensive overview is provided of the three principal components contained in the process of air pollution: meteorological processes, atmospheric chemistry and transformation processes, and the emissions sources. The specific research tasks to be carried out are defined. The contents include: Overview of Air Pollution Modeling; Meteorological Processes; Atmospheric Chemistry and Transformation Processes; Emission Estimates; Economic and Social Impact Studies; and Technology Transfer.

17. Key Words and Document Analysis. 17a. Descriptors

Air pollution	Photochemical reactions
Regions	Hydrocarbons
Meteorology	Nitrogen oxide
Chemical reactions	Oxidizers
Exhaust emissions	Particles
Sources	pH
Mathematical models	Carbon monoxide
Research projects	Planning
Atmospheric diffusion	Precipitation (meteorology)
Sulfur dioxide	

17b. Identifiers/Open-Ended Terms

17c. COSATI Field/Group 13B

18. Availability Statement
Unlimited

19. Security Class (This Report) UNCLASSIFIED

20. Security Class (This Page) UNCLASSIFIED

21. No. of Pages 284

22. Price

STANFORD RESEARCH INSTITUTE
Menlo Park, California 94025 · U.S.A.

Final Report *January 1972*

REGIONAL AIR POLLUTION STUDY: A PROSPECTUS

Part II — Research Plan

Prepared for:

THE ENVIRONMENTAL PROTECTION AGENCY
NATIONAL ENVIRONMENTAL RESEARCH CENTER
RESEARCH TRIANGLE PARK, NORTH CAROLINA

CONTRACT 68-02-0207

SRI Project 1365

R. T. H. COLLIS, *Director*
Atmospheric Sciences Laboratory (Project Director)

DON R. SCHEUCH, *Vice President*
Office of Research Operations

FOREWORD

This Prospectus was prepared by Stanford Research Institute for the Environmental Protection Agency under Contract No. 68-02-0207. While this Prospectus has been reviewed by the Environmental Protection Agency and approved for publication, approval does not signify that the contents necessarily reflect the views and policies of the Environmental Protection Agency, nor is it intended to describe the Agency's program.

The complete Prospectus for the Regional Air Pollution Study is presented in four parts.

Part I	Summary
Part II	Research Plan
Part III	Research Facility
Part IV	Management Plan

A table of contents for all parts is provided in each of the four parts to facilitate the use of the Prospectus.

ACKNOWLEDGMENT

This Prospectus was prepared at the Institute by a project team representing the full range of disciplines necessary for the comprehensive analysis of problems of air pollution. Research team members were drawn from four of the eight Institute Research Divisions, including the following:

Electronic and Radio Sciences
Physical Sciences
Information Science and Engineering
Engineering Systems

Because of the interdisciplinary nature of the effort, the contributions and research findings of many team members are distributed throughout this Prospectus rather than concentrated in one or more specific chapters. Accordingly, contributions are acknowledged below by general areas associated with the study of air pollution problems.

This Prospectus was prepared under the supervision of R.T.H. Collis, Project Director. The Project Leader was Elmer Robinson (now of Washington State University) until 15 January, when Richard B. Bothun, who had been Deputy Project Leader, succeeded him.

The main contributions were as follows:

- Elmer Robinson--Project leadership and the formulation of the Research Plan

- Richard B. Bothun--Project leadership and administrative management and the formulation of the Management Plan.

Technically, the principal contributions were:

- Richard B. Bothun--Management, scheduling, costing, planning

- Leonard A. Cavanagh--Air quality instruments, atmospheric chemistry

- Ronald T. H. Collis--Meteorology, remote sensing, research planning
- Walter F. Dabberdt--Transport and diffusion modeling, meteorology, instrumentation
- Paul A. Davis--Solar radiation, tracer studies
- Roy M. Endlich--Meteorological models, satellite systems
- James L. Mackin--Helicopter and aircraft systems
- Elmer Robinson--Meteorology, instrumentation, atmospheric chemistry, research planning
- Sylvin Rubin--Data processing systems
- Konrad T. Semrau--Source inventory and emissions
- Elmer B. Shapiro--Communication systems
- James H. Smith--Atmospheric chemical transformation processes
- Eldon J. Wiegman--Synoptic climatology

Valuable contributions were made in the latter stages of the project by Dr. W. A. Perkins and Mr. J. S. Sandberg, consultants.

The Institute wishes to express its appreciation for the assistance and provision of information by many staff members of the Environmental Protection Agency, especially Charles R. Hosler, Contracting Officer's Technical Representative; Dr. Warren B. Johnson, Jr., Chief, Model Development Branch; Robert A. McCormick, Director, Division of Meteorology; and Dr. A. P. Altshuller, Director, Division of Chemistry and Physics.

Additionally, the constructive criticism and comment provided by members of the Meteorology Advisory Committee of the Environmental Protection Agency during the preparation of the Prospectus were of significant value, and our indebtedness is hereby acknowledged.

CONTENTS

PART I - SUMMARY

FOREWORD iii

ACKNOWLEDGMENT v

I THE BASIC PREMISE 1

II SCIENTIFIC AIR QUALITY MANAGEMENT 3

The Basic Tool--The Mathematical Model 3
The Processes To Be Modeled 3
Accuracy 4
Current Limitations of Modeling 4
Steps To Improve Models 5
The Regional Scale 7

III THE REGIONAL AIR POLLUTION STUDY (RAPS) 9

Concept 9
Purpose 10
Organization 10
Objectives 14

IV SITE SELECTION 17

V THE RESEARCH PLAN 19

Introduction 19
Model Evaluation and Verification Program 21
  Meteorological Factors 24
  Pollutant Source Estimates 25
  Air Quality Measurements 25
Atmospheric, Chemical, and Biological Processes 26
Human Social and Economic Factors 28
RAPS Technology Transfer 28

CONTENTS

V Continued

Schedules and Task Specifications for the Research Plan . . 30
Introduction 30
100 Model Verification 45
101 Boundary Layer Meteorology Program 46
102 Emission Inventory 46
103 Air Quality Measurements 47
104 Model Calculation and Verification 47
200 Atmospheric, Chemical, and Biological Processes . . . 48
201 Gaseous Chemical Processes 49
202 Atmospheric Aerosol Processes 49
203 Other Pollutant Related Atmospheric Processes 51
204 Atmospheric Scavenging by Precipitation 51
205 Air Pollutant Scavenging by the Biosphere 52
206 Atmospheric Processes 53
300 Human, Social, and Economic Factors 54
301 Human and Social Factors 54
302 Economic Factors 55
400 Transfer of RAPS Technology for Control Agency Applications and the Formulation of Control Strategies . 55
401 Source Inventory Procedures 57
402 Atmospheric Monitoring 57
403 Data Handling 58
404 Modeling Technology 59
405 Other Significant Factors in Control Strategy Formulation 60

VI THE FACILITY 63

Rationale 63
Basic Operations 63
Basis for Monitoring Network 63
The St. Louis Regional Monitoring Network 66

CONTENTS

VII MANAGEMENT AND SCHEDULING 71

Introduction 71
Facility Activation Schedule 71
Permanent Management and Staffing 73

VIII COST SUMMARY 77

Permanent Facilities and Staff 77
Helicopter and Mixing Layer Observational Program 80
Research Plan 80
  Personnel 81
  Instrumentation and Equipment 83
  Operations 85
  Total Cost of Research Plan 86
Total Costs of RAPS 86

from C I-1

CONTENTS

The [illegible] includes:

PART II - RESEARCH PLAN

FOREWORD iii

ACKNOWLEDGMENT v

I INTRODUCTION TO THE RESEARCH PLAN I-1

II RESEARCH PLAN--OVERVIEW OF AIR POLLUTION MODELING II-1

  Introduction . . . . -1
  Model Evaluation and Verification Program . . . . -3

III RESEARCH PLAN--METEOROLOGICAL PROCESSES III-1

  Introduction . . . . -1
  Atmospheric Dispersion Models . . . . -4
    Gaussian Formulae . . . . -4
    Gradient Transfer Theory . . . . -11
    Other Models . . . . -24
  Numerical Weather Prediction Models . . . . -27
  Model Sensitivity to Meteorological Variables . . . . -30
  Experimental Meteorology Program . . . . -46
    General . . . . -46
    Tracer Studies of Transport and Diffusion . . . . -48
    Urban and Rural Radiation Budget Studies . . . . -62
    The Role of Remote Probing Techniques . . . . -67
    Upper Air Sampling Program . . . . -74
    Plume Dispersion Studies . . . . -79
    Studies of Spatial Variabilities . . . . -81
  References . . . . -84

IV RESEARCH PLAN--ATMOSPHERIC CHEMISTRY AND TRANSPORTATION PROCESSES, ... IV-1

  Introduction . . . . -1

to C xii

CONTENTS

IV Continued

The SO_2 Cycle IV-2
- Emission Sources of Sulfur Compounds . . . . -3
- Chemical Reactions of Importance to the SO_2-Cycle Model -5
- Removal Mechanisms for Sulfur Compounds . . . . -8
- The Research Program . . . . -9

The Photochemical Cycle--Hydrocarbons:Nitrogen Oxides: Oxidant . . . . -12
- Sources of Nitrogen Oxides . . . . -14
- Reactions of Nitrogen Oxides in the Absence of Hydrocarbons . . . . -15
- Hydrocarbon Sources and Removal Processes . . . . -17
- The Hydrocarbon-Nitrogen Oxides Reactions . . . . -18
- The Research Program . . . . -20

The Particulate Cycle . . . . -24
- Background Haze . . . . -25
- Natural Background Aerosols . . . . -27
- Particulate Emissions Inventory . . . . -28
- The Chemistry of Particulate Formation in the Atmosphere . . . . -30
- The Research Program . . . . -32

Development of Continuous Rainfall pH Measurement and Sequential Precipitation Collection . . . . -34

Carbon Monoxide Cycle . . . . -35
- Source of CO . . . . -37
- Important Chemical Reactions of CO . . . . -39
- CO Sinks . . . . -40
- The CO Research Program . . . . -41

The Chemical Research Program Schedule . . . . -42
- Aerosol Research Program . . . . -44
- The SO_2 Flux Measurement . . . . -46
- The $HC:NO_x$ Research Program . . . . -47
- The CO Research Program . . . . -51

References . . . . -53

CONTENTS

V RESEARCH PLAN--EMISSION ESTIMATES V-1

Requirements of the Emission Inventory System . . . . . . -1
Classification of Emission Sources . . . . -2
Source Processes . . . . -5
Source Units . . . . -5
  Stationary Sources . . . . -5
  Mobile Sources . . . . -7
Pollutants . . . . -7
Factors Affecting Emission Levels . . . . -8
  Stationary Sources . . . . -8
  Mobile Sources . . . . -10
Inventory Procedures and Accuracy . . . . -11
  Accuracy of Estimates . . . . -11
  Procedures . . . . -13
Emission Model . . . . -17
Control Strategy Studies . . . . -18
Inventory Schedule . . . . -18

VI RESEARCH PLAN--ECONOMIC AND SOCIAL IMPACT STUDIES VI-1

Introduction . . . . -1
Human and Social Factors . . . . -2
Economic Factors . . . . -5

and

VII RESEARCH PLAN--TECHNOLOGY TRANSFER VII-1

Introduction . . . . -1
Technology Transfer Program . . . . -1

VIII OTHER AGENCY RESEARCH PROGRAMS VIII-1

METROMEX . . . . -2
NCAR Fate of Pollutants Study (FAPS) . . . . -3
NOAA's MESOMEX . . . . -4

APPENDIX SCHEDULES AND TASK SPECIFICATIONS FOR THE RESEARCH PLAN A-1

CONTENTS

PART III - RESEARCH FACILITY

FOREWORD iii

ACKNOWLEDGMENT v

IX INTRODUCTION TO FACILITY DESIGN AND OPERATIONS IX-1

X ST. LOUIS SITE SELECTION X-1

Summary . . . . -1
Site Selection Criteria . . . . -4
Method of Analysis . . . . -7
National Summary Analysis . . . . -8
  Meteorology . . . . -8
  Fossil Fuels . . . . -9
  Regional Isolation . . . . -11
  Results . . . . -12
General Analysis of Standard Metropolitan Statistical Areas . . . . -12
  Pollutants . . . . -15
  Manufacturing . . . . -17
  Geographical Separation . . . . -21
  Sunshine . . . . -22
  Space Heating . . . . -22
  Results . . . . -23
  Agriculture . . . . -24

XI NEAR-SURFACE ATMOSPHERIC RESEARCH FACILITY XI-1

General Considerations . . . . -1
Horizontal Extent of the Network . . . . -1
Network Orientation . . . . -14
Characteristics of Stations . . . . -23
Meteorological Instrumentation . . . . -27

CONTENTS

XII AIR QUALITY SAMPLING XII-1

Introduction . . . . -1
Permanent Network Monitoring Stations for Pollutants . . -1
Semipermanent Monitoring Station . . . . -4
Pollutants To Be Monitored for Establishment of Air Quality . . . . -7
  Carbon Monoxide . . . . -8
  Methane, Nonmethane Hydrocarbons, and Total Hydrocarbons . . . . -8
  Nitrogen Oxides . . . . -8
  Sulfur Oxides and Hydrogen Sulfide and Total Sulfur . . -8
  Ozone . . . . -9
  Suspended Particulate Material . . . . -9
  Other Pollutants of Interest Not Measured by Network . -9
Pollutant Monitoring Techniques . . . . -10
  Carbon Monoxide, Methane, and Total Hydrocarbon . . . . -12
  Nitric Oxide, Total Nitrogen Oxides . . . . -13
  Sulfur Dioxide, Hydrogen Sulfide, Total Sulfur . . . . . -15
  Ozone . . . . -17
  Suspended Particulate Material . . . . -18
  Summary of Pollutant Instruments . . . . -20
Instrument Calibration . . . . -21
  Local Calibration . . . . -21
  Calibration Vans for Primary Calibration . . . . -23
Role of Research Programs . . . . -23

XIII DATA ACQUISITION AND HANDLING XIII-1

Introduction . . . . -1
Policies and Principles . . . . -1
System Overview . . . . -2
Instruments . . . . -6
Data Acquisition Equipment at Stations . . . . -8
Data Formats . . . . -13
The Central Data Collection Facility . . . . -15
  Overview . . . . -15
  Functional Tasks . . . . -16
  Equipment Complement . . . . -18

CONTENTS

XIII Continued

The Central Data Collection Facility (cont.)

Disk File Organization XIII-19
Manual Data Entry . . . . -20
Reliability Considerations and Maintenance . . . . . -20
Communications for the Data Collection Network . . . . -21
The Telephone Companies . . . . -21
Facilities . . . . -22
Costs . . . . -23
Alternative Communication Approaches . . . . -24

XIV MIXING LAYER OBSERVATION PROGRAM XIV-1

Introduction . . . . -1
General Mixing Layer Observation Methods . . . . -1
Primary Aircraft Support for the Regional Study . . . . -3
Helicopter Support Function . . . . -4
Schedule of Operations . . . . -4
Costs . . . . -9
Helicopter Instrumentation Package . . . . -9
Balloon Tracking System . . . . -14
System Design Concept . . . . -14
Activation Schedule . . . . -17
Costs . . . . -17
Special Aircraft-Based Meteorological Observations . . -18
General Concepts . . . . -18
Instrumentation Considerations . . . . -19
Special Aircraft-Based Air Quality Observations . . . . -24
Western Environmental Research Laboratory (WERL) . . . -24
Selection of Parameters . . . . -26
Selection of Surveillance Techniques . . . . -26

XV GENERAL CLIMATOLOGY OF ST. LOUIS XV-1

Introduction . . . . -1
Principal Seasonal Meteorological Characteristics . . . -3
Cold Season (Mid-October to Mid-April) . . . . -3
Warm Season (Mid-April to Mid-November) . . . . -8

CONTENTS

XV Continued

Meteorological Parameters Relating to Pollution XV-9
  Stagnating Anticyclones . . . . -9
  Wind . . . . -13

XVI LAND AND BUILDING REQUIREMENTS XVI-1

Central Facility . . . . -1
  Selection Criteria . . . . -1
  Interior Space Requirements . . . . -4
  Outdoor Facilities . . . . -6
Instrument Station Sites . . . . -7
  Selection Criteria . . . . -7
  Implementation . . . . -9

CONTENTS

PART IV - MANAGEMENT PLAN

FOREWORD iii

ACKNOWLEDGMENT v

XVII INTRODUCTION TO THE REGIONAL STUDY SCHEDULING, STAFFING, AND COST XVII-1

- Introduction . . . . -1
- The St. Louis Facility . . . . -2
- Facility Activation Schedule . . . . -5
- Permanent Management and Staffing . . . . -7
- Costs . . . . -9
  - Permanent Facility and Staff . . . . -9
  - Helicopter and Mixing Layer Observational Program. . . -12
  - Research Plan . . . . -12
  - Total Costs . . . . -14

XVIII IMPLEMENTATION SCHEDULE OF THE ST. LOUIS FACILITY . . . XVIII-1

- Introduction . . . . -1
- Prototype Instrument Station and Central Facility . . . -3
  - Schedule Network and Estimated Activity Durations . . -3
  - Activity Descriptions . . . . -17
  - Network Critical Path . . . . -27
- Activation Schedule of Class A and Class B Stations . . -33
  - Unit Schedules . . . . -33
  - Sequential Station Activation Schedule and Maintenance Requirements . . . . -36
- Full Facility Implementation . . . . -44
  - General Scheduling Conditions . . . . -44
  - Class A and B Station Activation with Prior Prototype Station Acceptance . . . . -53
  - Class A and B Station Activation Without Prior Prototype Station Acceptance . . . . -55
  - Class C Station Activation Schedule . . . . -59
  - Aggregate Facility Activation Schedule . . . . -63

CONTENTS

XIX PERMANENT MANAGEMENT AND STAFFING XIX-1

Introduction . . . . -1
Regional Study Management . . . . -1
Research Triangle Park Staff . . . . -6
St. Louis EPA Operating Staff with EPA Operation . . . -13
St. Louis EPA Staff with Prime Contractor Operation . . -25
St. Louis Facility Implementation . . . . -26
Staff Scheduling . . . . -32

XX ST. LOUIS FACILITY INITIAL COSTS AND ANNUAL OPERATING COSTS XX-1

Introduction . . . . -1
Initial Costs of the St. Louis Facility . . . . -1
  Air Quality and Meteorological Instruments . . . . . -3
  Instrument Station Preparation, Facilities, and Appurtenances . . . . -5
  Ditigal Data Terminal and Communication Equipment . . -6
  Central Facility and Equipment . . . . -9
  Vehicular Support Facilities . . . . -11
  Total Initial Costs . . . . -13
Annual Operating Costs . . . . -15
  Personnel . . . . -17
  Instrument Replacement and Spare Parts . . . . -17
  Telephone Communication System . . . . -19
  Motor Vehicles . . . . -20
  Building and Land Rental . . . . -21
  Miscellaneous . . . . -22
  Total Estimated Annual Operating Costs . . . . -23

XXI RESEARCH PLAN COSTS XXI-1

Introduction . . . . -1
Personnel . . . . -2
  Requirements . . . . -2
  Costs . . . . -5
Instrumentation and Equipment . . . . -6
Operations . . . . -10
Total Cost . . . . -11

ILLUSTRATIONS

PART II - RESEARCH PLAN

II-1 Model Verification Program II-5

III-1 Vertical Profiles of Wind and Eddy Diffusivity III-34

-2 Peak Surface Concentration as a Function of Distance from an Elevated Source . . . . -35

-3 Peak Surface Concentration as a Function of the Radial Distance from the Source . . . . -36

-4 Sources of Input Errors and Uncertainties in Variables Utilized in Air Quality Simulation Models . . . . -41

-5 Effect of Wind Direction on Concentrations Computed for the Washington, D.C., Camp Station . . . . -42

-6 Effect of Moving Receptor Point . . . . -44

-7 Normalized Concentration as a Function of Stability and Mixing Depth . . . . -45

-8 Hypothetical Lateral-Longitudinal and Vertical-Longitudinal Cross Sections Showing Envelopes for Plumes Generated Simultaneously by Identical Point Source at Two Elevations . . . . -54

-9 Hypothetical Lateral-Longitudinal and Vertical-Latitudinal Cross Sections Showing Envelopes for Plumes Generated Simultaneously by Identical Point Sources Displaced Crosswind Along Y-Axis . . . . -57

-10 Hypothetical Surface, Vertical-Longitudinal and Vertical-Latitudinal Cross Sections for Plumes Generated Simultaneously by Identical Point Sources at Different Upwind Ranges from Crosswind and Vertical Samplers at x_o . . . -58

-11 Program Elements of Tracer Experiments . . . . -63

VI-1 Interrelationship of Models Pertaining to Air Quality . VI-3

TABLES

PART II - RESEARCH PLAN

III-1 Sensitivity Analysis of the Gifford and Hanna (1971) Dispersion Model III-32

-2 Calculated Values of the Mean and Standard Deviation of $[(\chi_T - \chi_E)/\chi_T]$ as a Function of Systematic Errors in the Lateral Diffusion Coefficient σ_Y, after Hilst . . -38

-3 Calculated Values of Mean and Standard Deviation of $(\chi_T - \chi_E)/\chi_T$ as Function of Systematic Errors in Vertical Diffusion Coefficient σ_z, after Hilst . . . . . -39

-4 Calculated Values of Mean and Standard Deviation of $(\chi_T - \chi_E)/\chi_T$ as Function of Systematic Errors in Wind Direction (θ); Hilst . . . . -39

-5a Atmospheric Conditions . . . . -69

-5b Pollutants . . . . -70

-6 Vector Error of Wind (m/sec) at 5 km as a Function of Tracking System and Ratio (Q) of Mean Wind to Ascent Rate . . . . -77

IV-1 Chemical Reactions of Importance to the SO_2-Cycle . . . IV-4

-2 The 14-Step SAI Model . . . . -21

-3 Hydrocarbon Reactivity in NO Photooxidation . . . . . . -22

-4 Chemical Reactions of Importance to the Particulate Cycle . . . . -26

-5 Average Particulate Chemical Analyses ($\mu g/m^3$) . . . . . -27

-6 Particulate Emissions Inventory of St. Louis . . . . . . -29

-7 The Chemical Research Program . . . . -43

V-1 Classification of Sources for Emission Inventory V-3

Chapter I

INTRODUCTION TO THE RESEARCH PLAN

Part II of this Prospectus presents the Research Plan of the Regional Study. Two methods of presentation of the Research Plan are used. The first method provides a comprehensive overview is provided of the three principal components contained in the process of air pollution: meteorological processes, atmospheric chemistry and transformation processes, and the emission sources. The ~~second method defines~~ specific research tasks to be carried out ~~during the study~~. are defined.

In the first presentation, the general state of the art of these components is discussed, and the major problem areas on which the Regional Study should be most strongly focused are identified.

The considerations of meteorological processes in Chapters II and III cover their general involvement and effects on relationships of their magnitudes to air pollution and the manner in which these processes are treated in several mathematical models having a relatively advanced degree of development. Several of these models are likely to be the subject of detailed analysis during the Regional Study.

The discussion of atmospheric chemistry and transformation processes in Chapter IV covers the atmospheric pollutant cycles of the principal pollutants, including sulfur oxides, nitrogen oxides, hydrocarbons, particulate matter, photochemical oxidants, and carbon monoxide. The atmospheric cycles of most of these pollutants are not well understood, and recommendations are presented for improvement in the state of knowledge of these cycles in their most critical sectors. The function of both particulate material as a scavenger for many pollutants is addressed, along with the role of precipitation in removing both gaseous and particulate material from the atmosphere.

Chapter V presents the need for, and the procedures necessary to develop and maintain, a source inventory. A general classification scheme of a source inventory is presented that first divides the inventory into stationary and mobile sources. These categories are divided, respectively, into area and point sources and area and line sources. A further breakdown for all is made into combustion and noncombustion sources. The concepts and general rules by which sources can be assigned to these groups are provided and the methods of data acquisition are treated.

Whereas the foregoing chapters of Part II treat the main scientific and technical aspects of the Regional Study, the latter chapters address important primary derivatives of the scientific effort. Chapter VI addresses companion research efforts that could be carried forward in the area of economic and social implications of air pollution and programs for its control and abatement. Air quality data acquired by the instrument system, the source inventory, and perhaps the scientific programs, together with independently gathered data on the area's population, land use, manufacturing operations, and other factors, should lay the base for sorely needed development and improved understanding of the economic and social problems entailed in air pollution.

Chapters VII and VIII generally cover the interaction of the Regional Study with other organizations. Chapter VII--Technology Transfer--advances the concept that the Regional Study will indeed include developments and advances in the state of the art of instrument systems, data handling and processing, model verification, and many other areas that can be profitably adapted for use in similar or quite different areas of research as well as operation. The general content of a research program to cover these activities is suggested.

Chapter VIII covers the need for the Regional Study to interact with a number of other ongoing research efforts in the St. Louis area. These efforts are described and anticipated points of interaction are identified. These seven chapters, as noted, provide an overview and discussion of the Research Plan.

The second presentation of the Research Plan is provided in the Appendix of Part II. This appendix consists of a set of specific research tasks drawn from the broader discussion, along with estimates of the scheduling and personnel requirements for each task. The tasks are placed within four principal groups and assigned a numerical identifier. The four principal groups are shown below.

100 Model verification

200 Atmospheric chemical and biological processes

300 Human, social, and economic factors

400 Transfer of RAPS technology.

Each group in turn is divided into between four and six components, with the 100 group, for example, divided into 101, 102, 103, and 104. Each of these are further divided into specific tasks using a decimal notation, 101.1, 101.2, and so on. Scheduling and personnel estimates are shown at the decimal level.

Chapter II

RESEARCH PLAN--OVERVIEW OF AIR POLLUTION MODELING

Introduction

The scope of research efforts in the field of boundary layer simulation modeling centers on the need to understand, describe, predict, and ultimately control air quality in the lowermost stratum of the atmosphere. Simulation modeling provides the necessary link between inferences gleaned from air quality data obtained at isolated, single-point monitoring stations and the broad, yet detailed, picture of air quality that is required over an entire urban region; it also permits assessment of the ramifications of actual or projected growth (zoning) patterns and emission control (proportional versus selective) procedures. Neiburger (1969) presents a detailed discussion of the application of modeling in the study and control of air pollution.

Simulation modeling of the boundary layer refers precisely to physical or mathematical modeling of the atmospheric planetary boundary layer--the lowermost stratum of the atmosphere (on the order of 1 km depth) in which the effect of surface friction on the wind field is manifested. From a more practical standpoint, it is also the layer in which atmospheric pollutants are generally emitted, transported, diffused, and transformed. Mathematical models include, generally, gradient-transfer, similarity, Gaussian plume and puff, and statistical formulations, while physical modeling is done with the aid of wind or water tunnels. The utility of the mathematical models lies in their ability to describe and parameterize the physics of the problem over a large region and to predict meteorological and air quality changes that may occur either in time (as a result of the progression or development of weather systems) or from the alteration of emission patterns. Ideally, mathematical models are capable of achieving arbitrary degrees of temporal and spatial resolution to solve specific problems. High resolution, for example, may be necessary when considering pollutant concentration in urban core areas, whereas relatively low spatial resolution may be required for the study on the mesoclimatic scale. Physical models are perhaps less flexible, yet are

---

Neiburger, M., Bull. Amer. Meteorol. Soc., 50(12), 957-965 (1969).

ideal for evaluating the gross features of, for example, pollutant distributions in extremely inhomogeneous or complex locations.

It is highly desirable that the RAPS program evaluate the ability of the more promising models to simulate the atmospheric environment on both the micro- and mesoscales. In this regard, the models should be evaluated according to the specific function that they may serve. Specifically, evaluation programs are recommended for the following three functional model types: (1) diagnostic, (2) predictive, and (3) climatic. The primary emphasis at this time should be placed on diagnostic (descriptive) models, because the current or steady-state distribution of pollutants on the mesoscale must be described before detailed prognostic models can be developed. Prognostic or predictive models in this sense do not include diagnostic models which may be input with anticipated meteorological and emissions data to simulate an expected condition. Rather, predictive models use current initial conditions to predict (on the order, say, of one or two days) meteorological and perhaps emission fields, thereby predicting the level of air quality for some time in the future. Moreover, emphasis must be given to the development and evaluation of dynamic climatological models which have the ability to describe the mesoclimate and changes that may result from mesoscale urbanization.

Toward the achievement of these goals, it is essential that the various models be evaluated (and refined, as appropriate) with "real" data collected in the field. Such data collection will further our ability to understand and simulate the lower atmosphere on the mesoscale (on the order of 250 km). But unless one is able to observe the process he is attempting to describe, simulation modeling may be little more than an esoteric exercise. A vast amount of effort has been expended in the development of the various mathematical models and in their subsequent evaluation. In almost no case have the observed data been obtained on a scale compatible with the resolution of the model computation. Virtually all meteorological and air quality data collected on a routine basis for purposes other than model verification are the result of single point measurements (or, at the very best, several adjacent points). As such, the observation is a representation of very localized conditions and, in perspective, is a measure of the integrated effect of various scales of motion: micro, meso, and macro. In many cases, it is the microscale phenomena that predominate and there can be little surprise at the inability of numerical models to simulate the observation when the model may, in fact, predict only average conditions over a broad area, say a one kilometer square or larger. Another objective of the program should therefore be the definition of the spatial variability of ambient air quality as well as the spatial resolution of the simulation models. In practice, a feedback between observations and computations should result whereby the observations define the appropriate time and space scales that the models need to achieve, and eventually the models are used to describe spatial and temporal variations on the basis of a few representative measurements.

Federal air quality guidelines define levels of air quality on several time scales: hourly, eight-hourly, daily, and annually. On the basis of results from the observation/modeling programs, spatial criteria may also need to be established. It appears equally necessary to define criteria where the air quality is evaluated in parallel, over various length (or area or volume) and time scales. In this regard and in consideration of the requirements of model verification, the data collection program must also concern itself with the potential of remote (long-path) observation techniques where a particular contaminant can be measured on the appropriate spatial scale.

In summary, an extensive air monitoring network is required to define the scope of the problem and to evaluate and refine the mathematical models that are to simulate the level of air quality over a broad region. As such, the network should be not only extensive, but also flexible so that it will serve the many purposes of the simulation program. It must be capable of providing data from the substreet microscale to the regional mesoscale. The observations will also be used to evaluate these models on a variety of temporal scales: simulation of existing conditions (diagnostic requirement), prediction of short-term changes (prognostic requirement), and evaluation of potential mesoclimatic alterations (extended prognosis). Physical modeling techniques should be employed to supplement the field observation program in urban core areas where the complexities of building structures may severely limit routine in situ measurements on a practical basis.

Model Evaluation and Verification Program

The evaluation of the various simulation models can be accomplished most efficiently by considering the models in terms of the functions that they may be expected to serve. For convenience, these functions may be divided into three parallel types: (1) the time frame and resolution of the model, (2) the spatial frame and resolution, and (3) the class of contaminants to be considered. The models may therefore be classified according to whether they simulate the quality of the air in terms of the concentration distribution of inert, reactive, or particulate contaminants over either localized or expansive regions for current or forecasted conditions. Accordingly, there are on the order of a dozen functions that a model or models may be expected to fulfill; that is, there may be localized, diagnostic models for inert pollutants which have application in planning and evaluation studies (such as in the study of expected carbon monoxide concentrations resulting from highway development) or regional, prognostic models of reactive contaminants required for emissions control procedures. Figure II-1 illustrates this concept, as well as the subsequent steps in a model evaluation and verification program. It may be further desirable to test the various models under a variety of distinct meteorological conditions. One such stratification could be the separation of low and moderate-to-high wind speed cases (isolation of stagnation conditions); other distinctions may be made between near steady-state and strong advective conditions, or strong versus weak insolation.

At this point, the purpose of the simulation will have been defined and certain forcing physical criteria established; specific models falling within this framework can then be introduced for evaluation and subsequent verification. It is strongly recommended that initially the performance of the models be evaluated against both observed measures of air quality and the predictions of a simple standard (or reference) model or models. The simple model used by Hanna (1971) or a relatively simple box or Gaussian formulation (see Chapter III) should be considered. Quantitative, statistical techniques should then be applied to test the model against the reference and the observations. If the test model cannot show significant superiority to the reference, then it can be safely excluded from the later, detailed evaluation program. Emphasis should also be placed on a qualitative assessment of the extent and nature of the required input data. If a given model performs well, but requires input data that may not be readily available at the present or in the foreseeable future, then alternative formulations or parameterization may be necessary for further consideration.

Having initially demonstrated its feasibility, the model should be evaluated in detail in order to define both its strong and weak points so that refinements may be made. This detailed model evaluation program should include both fundamental and applied research tasks for the testing of the basic components of the model; namely, meteorological, emissions, and transformation processes. The applied tasks are, in effect, individual evaluation programs for the three process areas (or submodels). Submodel predictions of wind, turbulence, stability, emissions, plume rise, reaction rates, and so forth would be compared with routine observations from the research network (which may not be strictly routine for nonresearch programs). The sensitivity of each model and submodel should be evaluated

---

Hanna, S. R., "Simple Methods of Calculating Dispersion from Urban Area Sources," paper presented at the Conference on Air Pollution Meteorology, Raleigh, N.C., Sponsored by Amer. Met. Soc., April, 1971.

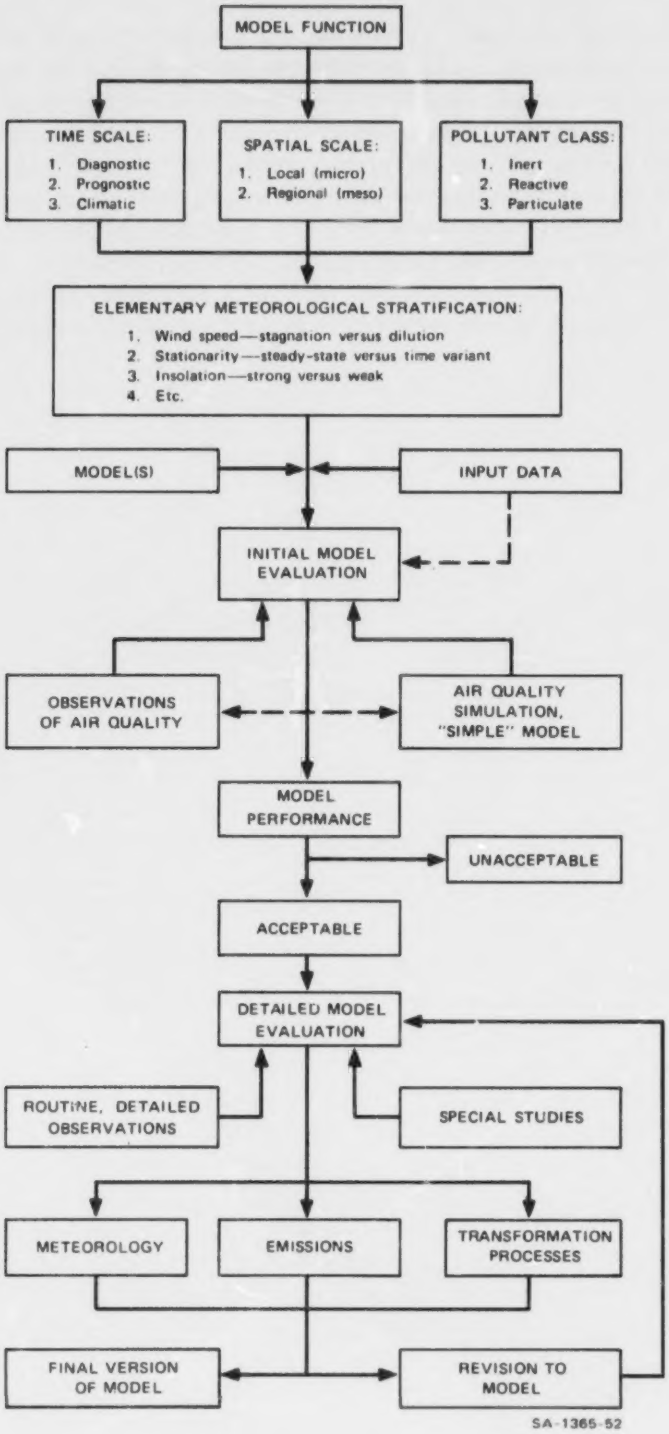


FIGURE II-1 MODEL VERIFICATION PROGRAM

with regard to the response of the output (model prediction) to variations of the input parameters. In addition to these direct or applied tasks, there is an acknowledged need for complementary research programs of a more fundamental nature. These would be directed toward providing a better understanding of the physical nature of the various physical processes so that the models may be revised accordingly and in conjunction with the requirements resulting from the applied program. Many of these fundamental programs can be anticipated; others, however, will result only as output requirements of the initial, detailed evaluations. The fundamental programs or special studies are given in detail in later sections.

Chapter III

RESEARCH PLAN--METEOROLOGICAL PROCESSES

Introduction

Most mathematical air quality simulation models may be classified conveniently in four major categories according to atmospheric dispersion theory: (1) gradient-transfer, (2) Gaussian (puff and plume), (3) similarity, and (4) statistical. In each type, the procedure is generally similar. An emissions field is specified (strength and spatial distribution), and then the field is redistributed and transformed according to the transport and diffusion characteristics of the atmosphere together with the consideration of reactions or deposition of the various contaminants. The manner in which the emissions, transport, diffusion, and transformations are specified varies a great deal among the various model types and indeed also among specific models of a given type. Perhaps the most important and certainly the most striking difference is in the treatment of diffusion.

The gradient-transfer method has as its basis the analog with the classical Fickian-type diffusion where the flux of mass or momentum is taken to be directly proportional to the gradient and where the factor of proportionality is the so-called eddy diffusivity. The simpler gradient models assume diffusivity profiles, while others either compute the variation on the basis of observations or through parameterization techniques. Diffusion in the Gaussian models is given by lateral and vertical standard deviation functions; these are usually proportional to travel distance from the source and the degree of atmospheric stability. Similarity models treat diffusion as occurring with a finite propagation rate determined from the characteristic turbulent length scale of similarity theory and are strictly applicable only in the atmospheric surface layer--that stratum adjacent to the earth's surface (on the order of 10 m depth) where height variations of the turbulent fluxes of mass, momentum, heat, or water vapor (as examples) are negligible in comparison with the magnitude of the surface flux and in consideration of measurement inaccuracies. In statistical theory, the diffusion is determined from consideration of the root-mean-square Lagrangian velocity fluctuations (measured, however, at a fixed point) and a characteristic autocorrelation function, usually under the assumptions of homogeneity and stationarity of the flow properties and coincidence between mass and momentum transport.

The essential objective of an evaluation and validation program for any of the various model types should be the development of a suitable model or models that will simulate meteorological and/or air quality conditions that are representative: (1) over a specified averaging period, (2) with the necessary spatial resolution, and (3) for the desired time frame, i.e., current conditions or predicted (short or long term) changes.

Less tangible objectives of the program also must be considered. The applicability of the various models must be evaluated in terms of both the immediate and future ability of air pollution control and planning personnel to utilize them. In this regard, the capability and accessibility of the required computational facilities must be evaluated. One must also weigh the input requirements of the various models in terms of the type of meteorological and emissions data that are currently available or can be anticipated. Compromises may be necessary to produce useful, workable, and tested models within the RAPS program. Attention must also be devoted to the more sophisticated and complex models that may be less suited for day-to-day operation but that serve a real need in the evaluation of the simpler models and in planning and impact studies. Also, techniques that will be required to process and analyze both measured and computed data must not be overlooked.

The large volume of data will require computer processing for all routine operations. Fortunately, there have been major developments in meteorological objective analysis that may be applied to this problem. Typical approaches to objective analysis have been given by Cressman (1959), Sasaki (1971), Stephens (1970), and Endlich and Mancuso (1968). In general, grid point values of a property of interest are determined from the measured values at surrounding stations, with higher weighting given to observations closer to the grid point. Variable importance may be assigned to different types of data, depending on sizes of random observational errors or other factors.

The general tendency of atmospheric properties to arrange themselves along the flow direction is included in the weighting scheme of Endlich and Mancuso. Stations are given a higher weighting if they are upwind or downwind than if they are crosswind at the same distance from the grid point. The determination of the flow direction is based on the observed wind directions. All these analysis techniques were derived for synoptic scale phenomena, but with suitable modifications they can be applied to regional boundary layer observations. Determinations of optimal weighting functions for regional analysis and other detailed matters can be made from the various data obtained early in the observational program.

The objective analysis methods currently in use are sufficiently flexible that they can be used with a variety of grids. For example, in using observed data spaced more densely in a central area in the manner discussed in Chapter XI, more than one grid spacing may be desirable. A fine-scale square grid could be used in the central area, surrounded by a grid of doubled spacing, with a third even larger grid near the extremity of the area of interest. Alternatively, analyses could be made on a grid of regularly increasing spacing away from a central point. These decisions on grids can be made in concert with decisions concerning the diffusion models that will be applied to air quality properties. It is worth noting also that analysis rules for air quality properties (such as ozone concentration) probably can be quite similar to those used for meteorological properties, since meteorological factors tend to control the dispersion of pollutants.

Meteorological quantities that would be analyzed include wind components (u, v, w), temperature, humidity, pressure, cloud cover and type, precipitation, and incoming and outgoing radiation. From these properties, other important factors would be calculated such as mixing layer depth and Richardson number. Sources of data would include the towers, surface stations, and upper-air facility of the regional network, surface and rawinsonde reports from standard networks, satellite cloud pictures in the visible and infrared, and radar measurements of precipitation. Satellite data with one-half mile resolution will probably be obtainable via telephone lines from a satellite readout station, which we understand is planned for installation at Kansas City by NOAA. Other standard data can be obtained in conventional ways.

From an objective analysis, values of any property can be obtained at any desired point by simple interpolation schemes. However, it is important to realize that the accuracy of the analyzed values (and interpolated ones) depends on the data coverage in that immediate vicinity. The highest reliability exists at the place and time of the greatest concentration of observations, regardless of whether analyses are done by hand or by computer.

The computer that assembles, processes, and analyzes these data should also be used to control one or more display devices. Rapid display of analyses will permit operational and research personnel to assimilate and assess changing events with minimal delay. This will be necessary eventually for real-time control of air quality. Consideration should be given to cathode-ray-tube displays; automatic line drawing equipment; video disk storage of maps, charts, and satellite pictures; and the software needed to operate these devices.

The various applications of objective analyses could be used in:

(1) Diffusion and air quality models and computations.

(2) Improved descriptions of small scale meteorological variables using spectral analysis techniques, leading to better objective analysis methods.

(3) Comparison of observed and predicted profiles of meteorological properties.

(4) Determination of trade-offs between space and time aspects of meteorological observations.

(5) Description of mesophenomena associated with cities such as the heat island effect and local thunderstorm occurrences.

(6) Research on boundary layer dynamics.

(7) Comparison of analyzed meteorological properties to cloud behavior shown by satellite data.

(8) Delineation of initial conditions for fine-mesh prediction models for the air quality control region.

The following sections will serve to present the structure, input requirements, and potential of several model types that appear capable of meeting some of the requirements stated earlier in this discussion.

Atmospheric Dispersion Models

Gaussian Formulae

Introduction

The most widely used simulation model at the current time is based on the Gaussian diffusion formulation. It is assumed that the downwind distribution of pollutant concentration (χ, M L^{-3}) from individual, instantaneous point sources of strength Q(M) is Gaussian-shaped in all directions,

$$\chi\,(x,y,z,t) = \frac{Q}{(2\pi)^{3/2}\,\sigma_x\sigma_y\sigma_z}\ \exp\left\{-\frac{1}{2}\left[\left(\frac{x-\bar{u}t}{\sigma_x}\right)^2 + \left(\frac{y}{\sigma_y}\right)^2\right]\right\}$$

$$\cdot\left\{\exp\left[-\frac{1}{2}\left(\frac{z+h}{\sigma_z}\right)^2\right] + \exp\left[-\frac{1}{2}\left(\frac{z-h}{\sigma_z}\right)^2\right]\right\}\ , \qquad \text{(III-1)}$$

where x is distance along the mean wind direction from the source (height h) and y is the horizontal and z the vertical crosswind directions. The variances (σ^2, L^2) are usually specified as a function of meteorological conditions and travel distance. Equation (III-1) is termed the Gaussian "puff" formula. If steady-state conditions are assumed, the time integration of the puff equation results in the so-called plume formula, where

$$\chi(x,y,z) = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}}\ \exp\left[-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2\right]\left\{\exp\left[-\frac{1}{2}\left(\frac{z-h}{\sigma_z}\right)^2\right]\right.$$

$$\left. + \exp\left[-\frac{1}{2}\left(\frac{z+h}{\sigma_z}\right)^2\right]\right\}\ . \qquad \text{(III-2)}$$

The height h is treated more appropriately as an effective source height where both the physical height of the emitter and the rise of the plume must be considered. Briggs (1969) has given a definitive analysis of the treatment of the effects of efflux velocity and buoyancy.

Ostensibly, the puff formula is more rigorous, since it can allow for the inclusion of departures of the flow field from the usually assumed straight motion, as well as for temporal variations of the emissions, transport, and diffusion characteristics. However, this is accomplished with a substantial increase in computation time and expense and with questionable improvement of the results. Pasquill (1971) has aptly pointed out that acceptance of the superiority of the puff method implies an ability to adequately describe trajectories and the growth of individual elements. However, the wide variability of the wind field over the spatial scale of the calculations suggests the use of an ensemble average, which is equivalent to the plume method. Previous studies (Islitzer and Slade, 1968) have shown that the assumptions explicit in the plume formulation are satisfactory for averaging times on the order of one-half hour and longer, although temporal changes of the mesoscale wind field may limit application for the longer averaging periods.

The basic Gaussian plume formulation can be applied to both point and area sources; in practice, the usual procedure is to represent an area source by an effective uniform emission strength and height and to consider anomalous sources by the addition of specific point source calculations. Various numerical integration techniques for the representation of the continuous area source have been evaluated by Shieh and Halpern (1971), while an analytical method has been used, for example, by Stanford Research Institute (Ludwig et al., 1970).

The SRI Model--an Example

The SRI model can be used conveniently to discuss Gaussian models in general; unlike some of the more extensive formulations that may be expected to be evaluated in the Regional Study (see, for example, the numerous Gaussian models presented at the Symposium on Multiple-Source Urban Diffusion Models [Stern, 1970] and the Conference on Air Pollution Meteorology [American Meteorological Society, 1971]), the SRI model has been well tested in an extensive field program (Johnson et al., 1971). Unlike other evaluation programs, the field experiment was specifically designed to collect data that would test all components of the model. As such, it did not suffer the limitations imposed by relying on available data--usually collected for some purpose other than model evaluation.

The fundamental theme of the SRI model is the simulation of the distribution of chemically inert, vehicle-generated pollutants in an urban area from existing or easily obtainable meteorological and emissions data. The model can be used in two ways: (1) in the synoptic mode, in which hourly concentrations are calculated from meteorological and traffic data; and (2) in the climatological mode, in which concentration frequency distributions are calculated on the basis of long term sequences of input data. The only input data required are airport weather observations and city traffic information. Because wind is the only input that is directly measured, submodels have been developed to estimate the emissions, atmospheric stability, variances, and mixing depth from the measured quantities.

The model is receptor-oriented, and the concentration at that point is determined by the atmospheric conditions and emissions from logarithmically spaced angular segments aligned upwind of the receptor along the mean transport wind. The innermost sector extends from the receptor to 125 m (comparable to a city block); the four inner sectors have a width of 45° to allow for large initial dispersion, while the outer six sectors are 22.5° in width ($\pm 2\sigma$) in accord with Gifford's

(1961) findings of plume spread under slightly unstable conditions. Emissions are assumed uniform within each segment but may vary between segments. The smaller sectors near the receptor provide the higher resolution required for nearby sources.

Equation (III-2) is integrated over x and y to permit application on an area-source basis,

$$\chi = \frac{0.8}{\bar{u}} \sum_{i=1}^{N-1} \bar{Q}_i \left[\frac{x_{i+1}^{1-b_j} - x_i^{1-b_j}}{a_j(1 - b_j)} \right] , \qquad \text{(III-3)}$$

where i is the number of the segment (i = 1, N) and j is the stability class. The parameters a and b are functions of atmospheric stability and result from the formulation for the variation of σ_z, the vertical standard deviation of the pollutant distribution,

$$\left(\sigma_z\right)_j = a_j x^{b_j} \quad . \qquad \text{(III-4)}$$

Equation (III-4) is based on an examination of experimental data on vertical diffusion in urban areas (Pooler, 1966; McElroy and Pooler, 1968; and Leighton and Dittmar, 1952, 1953 a-e).

Model predictions were compared initially with measured data from Continuous Air Monitoring Program (CAMP) stations; calculated and observed values often differed significantly in magnitude, although trends were similar. The failure of the model to predict observed values accurately resulted, in part, from the fact that the basic model treated only the diffusion process on the urban scale, while observations are greatly influenced by street-scale effects. A simple street effects submodel was subsequently developed and has greatly improved the performance of the model. The street effects submodel is based on "box-model" reasoning and predicts the contribution of very local sources to the total concentration as a function of roof-level wind direction, wind speed, street width, street traffic, and height and street side (e.g., windward, leeward). Field experiments were conducted in San Jose, California, and St. Louis, Missouri, for the purpose of model evaluation, refinement, and validation.

The SRI program serves to illustrate, in part, the basic nature and scope of the proposed RAPS program. The objectives of the SRI study were the refinement and validation of a practical, multipurpose regional (urban mesoscale to street microscale) dispersion model from the results of detailed field measurements. The measurement program was designed not only to evaluate the validity of the computed concentrations but also to test the degree to which the various model components (submodels) were able to simulate the input requirements of the basic model (in this case, Eq. [III-2]). The following discussion serves to expound on the nature of the input data requirements and their simulation to prescribe requirements that an observational network must satisfy for the total evaluation of Gaussian-type dispersion models.

The input requirements for virtually all classes of pollution simulation models may be classified conveniently into one of two types, emissions and meteorological. According to the manner in which the various models have been classed (i.e., by the nature of diffusion simulation), the problem of the input of emissions data is common to all models. The problem of the simulation of emissions may be further structured into three subelements: specification of the strength and spatial and temporal variations of the emissions field. The strength of emissions implies the fractionation into pollutant types as well as the rate with which they are emitted. Furthermore, modeling of the spatial and temporal variations must appreciate the nature of the emission-generating process together with the desired model resolution. For example, the modeling of daily concentrations of a pollutant may require specification of subday emissions when the basic emitting period is shorter than one day. The emissions problem is further complicated when considering mobile rather than stationary sources where both the strength and spatial distribution may be some temporal function.

In its model, SRI was concerned with the emission of carbon monoxide (CO) by automobiles. The strength of the emission of CO is a function of the number of vehicles and, more directly, of their speed and age. San Jose has an extensive computer-based traffic monitoring network that provides detailed traffic information for the central business district (CBD). Spatial and temporal distributions in the outer region are obtained from historical traffic data. With the available traffic information, evaluation of the emissions submodel was reduced to a check of the empirical equation that quantified the effect of vehicle speed and age. The evaluation of the emission of CO from the CBD was made from a CO mass budget analysis using surface (van) and aerial (helicopter) measurements around the area. The difficulties of effectively evaluating the simulation of a complex emission field--such as CO, SO_2, and NO--are large and accentuate the need for a major program of emission inventory design and simulation; the SRI verification program resulted in the conclusion that its submodel was not invalid. Although only a qualified success, its verification program is a pioneering effort for area source emissions modeling. Additional work in this important area is required and will most probably include the use and development of suitable, remote monitoring systems; a discussion of the task of emissions modeling is given in Chapter V.

Meteorological inputs to the model are required to describe atmospheric transport and diffusion; for the Gaussian models, this is done by the mean wind and the variance of the vertical pollutant distribution (see Eq. [III-2] or [III-3]). A time-averaged, low-level wind speed and direction are the only routine observations that may be used directly in the model. No definitive guidelines exist for the height at which this measurement should be made; in practice, available winds are often used with little regard for the height, location, sensor-type, and other factors. However, the results may not reflect these inaccuracies because of similar limitations in the emissions, diffusion, and concentration (observed) values, as well as the empirical nature of the model itself. Smith and Singer (1966) provide insight into the interpretation and use of the term in the equation. It is an indicator of the mean speed and direction with which dispersion is occurring. Because of the horizontal transport and vertical diffusion of a plume, it will encounter changes in the mean wind speed and direction. This effect is usually most pronounced as the plume grows vertically because of the well-known variation of wind with height, but it can also occur in the horizontal plane because of either temporal changes or horizontal variations in the wind field. To incorporate these effects in the Gaussian models, they suggest the use of a variable "mean equivalent wind" corresponding to the speed at the height of the vertical center of mass of the plume.

There are five basic methods for estimating the lateral and vertical standard deviation functions required in the Gaussian-type formulations; the development and evaluation of the various methods have been summarized by Slade (1968). The first method was developed by Sutton (1953) and follows from dimensional analysis of the proposed dependence of the Lagrangian single-particle autocorrelation function on only the intensity of turbulence and the eddy viscosity. The standard deviation functions can be estimated analytically from a knowledge of the turbulence intensity and the shape of the wind profile. Because of the widespread use of the Sutton formulation, algorithms have been developed to estimate his so-called virtual diffusion coefficients (which are proportional to σ_y and σ_z) from available observations of: (1) the shape of the wind profile or (2) magnitude of the wind speed together with an estimate of atmospheric stability. A second method for estimating the lateral and vertical standard deviation functions is through their functional relation to the lateral and vertical eddy diffusion coefficients. The application is somewhat questionable because of the practical (and theoretical) difficulties in determining the eddy diffusivity and in treating its height variation in view of the constancy of the standard deviation functions. The last three methods are more attractive (as evidenced by their popularity), since they parameterize the sigma functions on the basis of directly measurable atmospheric values. The method of Cramer (1957) relates σ_z and σ_y to the standard deviation of wind direction fluctuations and travel distance. Hay and Pasquill (1959) and Pasquill (1961) also estimate vertical and lateral spread from wind-direction fluctuation measurements. They further present, together with Gifford (1961), a parameterization technique for estimating stability and hence σ_z and σ_y from wind speed and insolation data, together with travel distance. Smith and Singer (1966) and Singer and Smith (1953) relate the standard deviation functions to wind direction spread (gustiness), as well as to travel distance in a form similar to Equation (III-4). Their gustiness classification is well known and inherently incorporates the effects of stability and wind speed.

The SRI model uses a modified form of the Hay and Pasquill method. During the San Jose program, the stability algorithm was evaluated (and slightly modified) on the basis of a comparison with a bulk stability coefficient computed from measured vertical profiles of temperature and wind. The dependence of σ_z on travel distance (x) and stability was also revised on the basis of recently available urban diffusion data (Leighton and Dittmar, 1952, 1953 a-e). The data for the Pasquill-Gifford formulations were based on measurements taken in England over rolling, wooded countryside containing small towns and so may not be strictly applicable for use in urban areas. Another question that therefore must be answered is the degree and nature of the dependence of σ_y and σ_z on the aerodynamic roughness of the interface boundary. The recent SRI field program in St. Louis was directed toward providing additional information in this area. Continuous measurements of wind direction fluctuations and vertical profiles of wind and temperature (for the computation of stability coefficients) were made in the urban center; results of the program are not yet available.

The SRI model also treats the effects of a limited mixing depth due to the presence of elevated inversions. Under these conditions, pollutants tend to become uniformally distributed in the vertical after sufficient travel time has taken place. Therefore, the model reverts to a simple "box" type when the surface concentrations of the two formulations first become equal. The afternoon (maximum) mixing depth is computed from the morning sounding of the nearest radiosonde station and is taken as the height where the potential temperature of the sounding is equal to the afternoon maximum surface potential temperature. Estimation of the nighttime mixing depth over a city is based on Summers' (1966) model of the urban heat island and an empirical relationship (Ludwig, 1968, 1970) between rural lapse rates and the intensity of the heat island effect. Hourly mixing depths are estimated by interpolation based on surface temperature during the day and on time for the evening. Evaluation of the mixing depth submodel was based on helicopter temperature profiles and the lidar, remote probing technique.

Other investigators have examined the problem of treating the effect of a limiting mixing depth in the Gaussian model. Meade (1959) suggests the cessation of vertical spreading when $\sigma_z = 0.456$ H, but this has the undesirable feature of a substantial vertical concentration gradient below the lid. Pasquill (1962) has proposed that σ_z should become a constant after reaching H, and is based on the desire to maintain a near-constant concentration profile at this point. Smith and Singer (1966) have suggested what is essentially the procedure used in the SRI model; they have proposed that $\sigma_z = 0.8$ H in the limit with a lid such that the ground level concentration given by their model would be consistent with the value for a uniform vertical distribution (box model).

Finally, the effects of pollutant transformation and removal processes must be considered in the model. This includes the deposition of particles and gases, the effects of precipitation scavenging, and the transformations of chemical and photochemical reactions. These are common to all model types and are discussed at greater length in Chapter IV.

In summary, the direct atmospheric inputs to the Gaussian-type models are the effective transport wind speed and direction, mixing depth, and standard deviation (diffusion) functions. None of these parameters are either easily or routinely measurable, and they require submodels for their evaluation. Various formulations have been discussed, and the SRI model has been presented to illustrate the structure of a Gaussian model, together with the type of field program that is necessary for its evaluation. Specific research tasks required for the refinement and evaluation of Gaussian (and other) models are presented later in this chapter.

Gradient Transfer Theory

General

The gradient transfer or so-called K-theory is based on the solution of the equation for the diffusion of mass postulated by Fick, in analogy with similar governing equations of other disciplines: Darcy's law for groundwater flow, Fourier's law for heat conduction, Maxwell's law of electrostatics, and Ohm's law for current conduction. The classical form for these equations is

$$\frac{d\bar{q}}{dt} = K \frac{\partial^2 \bar{q}}{\partial x^2} \quad , \tag{III-5}$$

where $\bar{q}$ is the mean value of the particular variable and K is a constant diffusion coefficient--the eddy diffusivity for the atmospheric case. Equation (III-5) can be reexpressed in a more general form where the diffusion is treated in three dimensions and the coefficients are not necessarily equal and can vary in space,

$$\frac{d\bar{q}}{dt} = u \frac{\partial \bar{q}}{\partial x} + v \frac{\partial \bar{q}}{\partial y} + w \frac{\partial \bar{q}}{\partial z} + \frac{\partial \bar{q}}{\partial t} = \frac{\partial}{\partial x}\left(K_x \frac{\partial \bar{q}}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial \bar{q}}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z \frac{\partial \bar{q}}{\partial z}\right) + S_q \quad , \tag{III-6}$$

where S_q is the source/sink term of the parameter $\bar{q}$. It may be worthwhile at this time, in view of the discussion of the preceding section, to point out that the solution of Equation (III-5) for a stationary medium with a point source of $\bar{q}$ is a Gaussian function.

The applicability of the concept of an effective or eddy coefficient for the determination of the diffusion of mass, momentum, or energy in the atmosphere is open to some question. The eddy diffusivity for momentum has its physical basis in the mixing length hypothesis of Prandtl; its application to mass and energy diffusion may be conjectured, considering two points: (1) if the scale of the diffusive action is sufficiently small to permit representation by the mean gradient and (2) to what degree a diffusion coefficient for mass (or energy) can be determined from observed properties of the eddy (wind) field, as is the usual procedure. These points have been the subject of considerable discussion over the years, and Pasquill (1971) has presented a recent synopsis. The general acceptance of the practical utility of the method may be judged by the wealth of papers that treat the diffusion process in this manner (see, for example, the review given in Slade, 1968) and the many useful results achieved. A quotation attributed to Corrsin (1959) aptly sums up the situation: "... K theory is not useful in principle but only in practice."

In practice, solutions of various forms of the general equation (III-3)--commonly referred to as the conservation equation--usually follow three procedures for the treatment of the wind and diffusion terms. In the simplest case, a priori knowledge of the distribution of wind and diffusivity components is assumed known. A second and more general type specifies the wind field and then computes the diffusion coefficients from a suitable model relating flux and gradient. The third and ultimate model generates all dependent variables.

An alternative, yet related, classification scheme is through consideration of the various specific approaches used to solve the conservation equations. Generally, this latter approach classifies the models as either Eulerian (fixed coordinate system) or Lagrangian (moving-cell concept); Roth et al. (1971) have given a fairly comprehensive evaluation of models in these terms. Model differences are reflected primarily in the mathematics of the solutions, while the classification of the transport and diffusion processes is secondary.

The moving-cell approach traces the trajectory of a hypothetical air parcel across an airshed. The model assumes that the parcel maintains its integrity throughout its movement, thereby neglecting horizontal transport or diffusion across the lateral boundaries of the parcel and the height variation of the wind. This approach greatly simplifies the solution and provides an assessment of the effect of specific sources along the trajectory. Aside from the simplification of the atmospheric process, the results are limited to the given trajectory. The Eulerian (fixed coordinate) approach is subdivided into: (1) conventional finite difference solutions, (2) the so-called particle-in-cell (PIC) method, and (3) the well-mixed cell model. The PIC method is characterized by representing the continuous concentration field by a finite number of mass points. The third Eulerian method represents the airshed by a three-dimensional array of uniformly mixed cells where intercellular transport, but not diffusion, is permitted.

Roth, et al. (1971) conclude that the various models are only now reaching a state of development where direct comparisons may be made. In view of the various simplifications and mathematical approaches that the models encompass, it seems desirable to approach an assessment of K-theory modeling through examination of the most general type in terms of the treatment of the atmospheric process. A finite difference model of transport and diffusion in the urban planetary boundary layer by Pandolfo et al. (1971) perhaps best illustrates the current state of the art of regional K-theory modeling. As such, it will be used to illustrate some of the various features of the K-theory models with the objective of examining its potential for application and, subsequently, its evaluation and validation within the proposed RAPS program. Alternative formulations will be cited where appropriate.

A Complex Numerical Prediction Model

The model developed by Pandolfo and his associates (1971) at the Center for the Environment and Man (CEM) is a numerical integration of the K-theory forms of the conservation equations for mass, momentum, heat, and water vapor. The procedure incorporates generation of the wind and diffusivity fields from initial and boundary conditions and is generally applicable to any environment as a result of the parameterization of the various fluxes in terms of geophysical parameters (e.g., latitude, solar hour angle, albedo, aerodynamic roughness). Because of the simultaneous solution of the five conservation equations, feedback loops between, for example, the pollutant and temperature or wind fields can be considered.

The CEM model is in reality a family of models with different degrees of sophistication and objectives. The model can be applied in the predictive as well as the diagnostic mode and has been used to evaluate the three-dimensional structure of the mesoscale planetary boundary layer, as well as to analyze height and temporal variations at a single station.

When the equations of motion for the planetary boundary layer are considered together with the gradient transfer assumption, the equations for the conservation of horizontal momentum are given as

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial x} = \frac{\partial}{\partial z}\left(K_m \frac{\partial u}{\partial z}\right) + f(v - v_g) \quad , \qquad \text{(III-7)}$$

$$\frac{\partial v}{\partial t} + v\frac{\partial v}{\partial y} = \frac{\partial}{\partial z}\left(K_m \frac{\partial v}{\partial z}\right) - f(u - u_g) \quad , \qquad \text{(III-8)}$$

where u and v are the wind components (LT^{-1}) in x and y, respectively; the subscript g refers to the geostrophic value which represents the acceleration due to the horizontal pressure field, f is the Coriolis parameter (T^{-1}), and it is assumed $K_{u-z} = K_{v-z} = K_m$. The contribution by the terms for the horizontal diffusion of momentum can be assumed to be negligibly small in comparison with the vertical component, i.e.,

$$\left|\frac{\partial}{\partial z}\left(K_z \frac{\partial[u,v]}{\partial z}\right)\right| >> \left|\frac{\partial}{\partial x}\left(K_x \frac{\partial[u,v]}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial[u,v]}{\partial y}\right)\right| \quad .$$

Alternately, the effects of horizontal diffusion may be treated implicitly in the computational scheme.

The equation for the conservation of heat follows in a similar fashion from (III-6),

$$\frac{\partial T}{\partial t} + u\frac{\partial T}{\partial x} + v\frac{\partial T}{\partial y} + w\frac{\partial T}{\partial z} = \frac{\partial}{\partial z}\left(K_H \frac{\partial T}{\partial z}\right) + \Gamma \frac{\partial K_H}{\partial z} + \Gamma w + S_s + S_L \quad . \qquad \text{(III-9)}$$

K_H is the eddy diffusivity for heat, T is temperature (D), and Γ is the adiabatic lapse rate. The first term on the right-hand side of Equation (III-9) expresses the vertical diffusion of heat without the effects of adiabatic warming or cooling while the second and third terms describe the adiabatic effects resulting from turbulent and mean vertical exchanges, respectively. The last two terms describe the sources (or sinks) of heat due to solar (subscript s) and terrestrial (subscript L) radiation, respectively.

Moisture conservation is expressed by

$$\frac{\partial q^*}{\partial t} + u\frac{\partial q^*}{\partial x} + v\frac{\partial q^*}{\partial y} + w\frac{\partial q^*}{\partial z} = \frac{\partial}{\partial z}\left(K_W \frac{\partial q^*}{\partial z}\right) + S_W \quad , \qquad \text{(III-10)}$$

where q^* is the specific humidity (n.d.) and S_W describes the internal sources or sinks of moisture.

Finally, the mass conservation equations for the various pollutants are of the form

$$\frac{\partial P_i}{\partial t} + u\frac{\partial P_i}{\partial x} + v\frac{\partial P_i}{\partial y} + w\frac{\partial P_i}{\partial z} = \frac{\partial}{\partial z}\left(K_m \frac{\partial P_i}{\partial z}\right) + (S_p)_i + R_i \qquad \text{(III-11)}$$

and where P_i is the concentration (ML^{-3}) of the pollutant species, i, S_p represents the pollutant source or sink term ($ML^{-3}T^{-1}$), and R_i is the rate of generation or depletion ($ML^{-3}T^{-1}$) of species i by chemical reactions or fallout.

Exclusion of the horizontal diffusion terms cannot be as easily justified for the treatment of pollutant dispersion as it often can be for momentum, for example. However, proven quantitative formulations for horizontal diffusion in the planetary boundary layer are not available. Pandolfo (1971) goes on to state that, in view of this limitation, the relative accuracy of various finite difference schemes is rather academic at this time, provided the diffusive component of the inaccuracy is known. Therefore, the CEM model uses the diffusive computational error of the upwind differencing scheme to implicitly simulate the gross characteristics of the horizontal diffusion.

The simultaneous solution of Equations (III-7-11) which is based on a knowledge of the initial and boundary conditions, together with the diffusivity and source terms, would ideally permit both the description and prediction of the three-dimensional fields of mass, momentum, and energy within the limitations of computer technology and the applicability of the gradient-transfer theorem. The multitude of K-type dispersion models that have been formulated within the last decade are numerical solutions using varying simplifications and computational techniques of the general, composite problem as formulated above. For example, if the wind and diffusivity fields are specified rather than computed (see, as examples, Sklarew, 1970; and Seinfield, 1971), the dispersion model is reduced to the solution of Equation (III-11) alone. However, the procedure implicitly ignores the potentially significant feedback mechanisms between moisture, pollutants and temperature (and hence stability, wind, and diffusivity). In practice, the limitations imposed by these restrictions are not too severe, since the effects are likely second order.

However, use of the model is generally restricted to what conveniently may be classified as a planning or evaluation role where it is desired to examine the effects of: (1) a given or projected meteorological situation on the pollutant concentration field resulting from a specified emissions field or (2) conversely, a variable emissions field under given meteorological conditions.

There is indeed much to be said of the need for this particular application of the model. Aside from the practical requirements that have been cited, this form of the model provides a reliable test of the potential of the more complex (and expensive), interacting predictive models. For surely, if the simple model can only attain a certain degree of success in simulating a concentration field given an adequate set of meteorological conditions, then the potential of the complex model is similarly limited. Unfortunately, a completely adequate set of input meteorological data may only be available from extensive research programs such as the RAPS, and therefore the need arises to generate the necessary conditions from limited data, together with an appropriate procedure for extension through parameterization by readily available geophysical data. Along these lines, the research program must evaluate the parameterization scheme and specify the necessary spatial and temporal resolution (scales) that are required to model the physical atmospheric processes correctly.

The CEM model incorporates a horizontal grid mesh on the order of 10 x 10 with up to 30 levels; the finer vertical resolution is necessary to describe properly the large meteorological variations through the planetary boundary layer. The height variation of the eddy diffusivity is computed with the aid of the results of surface layer similarity theory. Although the surface layer is commonly considered as the shallow stratum adjacent to the earth's surface, it may be defined alternately as any layer in the planetary boundary layer where the height variation of the turbulent flux of momentum (or mass or heat) is negligibly small in comparison with the absolute value. The mean eddy diffusivity of heat and momentum thereby are computed for each layer from the slope of the wind, temperature, and moisture profiles through the gradient Richardson number and the stability regime, i.e., stable conditions, or forced or free convection. The vertical wind, temperature, and moisture profiles, in turn, are computed simultaneously. The enormity of the computational task is readily apparent on consideration of the fact that there are 17 independent variables (for the single pollutant case) in Equations (III-7-11). Therefore, additional independent equations (or submodels) are required to specify all the variables.

A thorough evaluation of a model of this type would require the examination of the performance of each of the submodels. The formulations for the diffusivities and the equations for the atmospheric heat source and sink terms due to solar and terrestrial radiative exchange in the presence of various particulates and gases are two specific cases where such evaluation is necessary. In short, evaluation would ideally require information on the three-dimensional and temporal structure of the distribution of mass, momentum, and energy in the planetary boundary layer on a regional scale.

Aside from the problems and complexities of realistically simulating the physics of the overall problem, the restrictions imposed by computer storage and computational time and expense must be considered. The latter two considerations can vary according to the type and purpose of the computations. A real time diagnostic or short term predictive model will require a minimal turnaround time, whereas use of the model for long range planning or evaluation purposes may impose little or no restriction on the time of the computation.

The limitation set by the finite storage capacity of the computer will implicitly affect the time problem, but more importantly, it will limit the spatial extent and/or resolution of the model. The CEM model, as an example, more than adequately provides the vertical resolution required but does not permit sufficient horizontal resolution for many purposes. The horizontal extent of a typical mesoscale region is on the order of 200 km for air quality considerations; the CEM model has been tested for the Connecticut area with a horizontal resolution of 24 km. This scale of resolution is probably adequate to describe the mesoscale features of the wind, temperature, and moisture fields over substantially homogeneous terrain, but it is not sufficient to model the effects of variations in the aerodynamic, thermal, and evaporative characteristics of the terrain that may arise from the presence of large towns and cities. The City of St. Louis occupies an area of 158 km^2 while the county is 1300 km^2. The problem of scaling is even more acute when considering the emissions field. Therefore, an objective of the RAPS should be the definition of the spatial (and temporal) scales appropriate to the various meteorological variables and pollutants. It may be necessary to consider nesting of the scales used in the solution of the model. For example, to simulate air quality conditions characteristic of a horizontal length scale of 1 km, it may be necessary to consider, say, the radiative and moisture fluxes on a 20 km scale, wind on the 5 to 10 km scale, and emissions on a scale of 1 km.

The CEM three-dimensional simulation test used historical winter data for the initial and boundary conditions. The grid extended over Long Island Sound and therefore tested the response of the model to the influence of extreme difference in surface conditions (topographic effects have not yet been incorporated in the CEM model). Pollution source data were obtained from the Connecticut source inventory of Hilst et al. (1967). Sulfur dioxide was the only pollutant considered in the test; unfortunately, chemical reactions have not yet been included in the model. The results really permit only a qualitative assessment of the model, but they do show the gross horizontal patterns expected from the varied (urban, rural, and water) surface characteristics. Systematic convergence patterns result over the urban areas and are reflected in the deeper mixing layers. Lower rural nocturnal temperatures are also predicted.

Although the three-dimensional form of the model is attractive in principle, it is not yet sufficiently developed to permit evaluation with existing or anticipated air quality data owing to weaknesses in the treatment of the following areas: (1) chemical and photochemical reactions, (2) horizontal advection, (3) topographic effects, and (4) horizontal resolution.

A more rigorous test of the model entailed the use of a single station, neglecting horizontal gradients and mean vertical motions. In this manner, the sensitivity of the model to a variety of external conditions was considered. Specifically, three single-station experiments were conducted: (1) estimation of the effects of surface characteristics on the interface temperature and atmospheric stability, (2) examination of the effects of varied source heights, and (3) examination of the effect of pollutants on the temperature structure and the feedback effects on the pollutant concentrations. The single-station model predicts realistic urban-rural differences of the near-surface temperature: winter daytime maxima are similar while the nocturnal minimum is 10°K lower for the rural case. The warmer urban nocturnal temperatures are attributed primarily to anthropogenic heating, with the effect of the thermal admittance of the substratum being secondary. The depth of the mixing layer is shown to be highly correlated with surface temperature; near-surface pollutant concentrations vary by a factor of five over the day with minimum concentrations occurring in the afternoon.

The effect of a fixed profile of pollutant concentration on the surface temperature was a 2°K decrease in both the amplitude and absolute value of the diurnal cycle. Another simulation evaluated the feedback between the pollutants and the temperature structure through the effect on the eddy diffusivity; the results projected a 25% change in pollutant concentration.

A Simple Model of Numerical Prediction

The numerical simulation model of Roth and his associates (1971) at Systems Applications (SA) is another example of a finite-difference solution to the gradient transfer form of the equation for mass conservation; the distribution (x, y, z, t) of the momentum and energy fields are specified on the basis of observations and empirical formulations. As such, the model is only prognostic in the sense that it predicts the ground level concentration field given a forecast of the horizontal wind field and vertical temperature profile. Unlike the CEM model, it does incorporate photochemical reactions and interactions of the contaminants as well as the effects of terrain irregularities. However, horizontal diffusion is ignored.

The SA model was tested initially to predict the distribution of carbon monoxide (CO) in the Los Angeles Basin. CO was chosen for the test, because it is inert and permitted the evaluation of the meteorological, emissions, and computational aspects of the model without the complexities of the reaction terms. The sensitivity of predictions of the average surface CO concentration for the two-mile square grid areas could be considered as a result of variations (both at the surface and aloft) in the: (1) wind field, (2) eddy diffusivity, (3) mixing depth, (4) computation scheme, (5) grid size, and (6) the integration time interval. Comparison of predicated concentrations with observations at 11 scattered monitoring stations shows generally good agreement for a 13-hour period of one test day (correlation coefficient of 0.65); a second period with light winds (and subsequently, high surface concentrations of CO) gave poorer results.

Results of the sensitivity tests are given only qualitatively. Roth et al. (1971) report that "modest" variations in the wind field and mixing depth have "significant" effects on the concentrations. Sensitivity to the vertical profile of diffusivity has not yet been evaluated. The finite difference method is accurate except when large concentration gradients are encountered. Finally, the emissions formulation has not been thoroughly evaluated, although considerable effort has been expended in obtaining an accurate traffic data base.

Emissions of CO in the Basin are mainly from automobiles (96%), with minor contributions from aircraft (3%). Automotive emissions are treated as a constant per vehicle-mile with higher values assigned to early morning hours because of "cold starts." Locally important emissions from power generating stations and refineries are also considered.

Specification of the temporal and spatial distribution of the mixing depth (H) over the Basin is based on the studies of the Los Angeles marine layer by Edinger (1959) and Edinger and Helvey (1961). Temporal variations are provided by 20 temperature profiles obtained over the two-day period at a total of four locations. The spatial distribution of H for 625 grid squares is given by Edinger's works. An evaluation of observed and predicted values showed good agreement for the morning hours and poor results in the afternoon.

The surface wind field was reconstructed from observations taken each hour at 34 locations by four different organizations. Streamline and isotach patterns were subjectively analyzed and then digitized over the grid. Little or no upper air wind data were available, and two gross assumptions were tested for the determination of the three-dimensional wind field. The first assumption considered the horizontal wind field as invariant with height; the second assumption specified the shape of the vertical profile of the vertical component of the wind. The vertical structure of the horizontal wind was then computed for the latter case, with certain restrictions placed on the magnitude of the wind speed and the amount of veering permitted.

The eddy diffusivity was not explicitly coupled to the wind field, but rather was specified empirically according to the work of Eschenroeder and Martinez (1969). According to their synthesis, the vertical profile of the diffusivity is given by the value of the surface wind speed and the height of the inversion lid where the diffusivity at the lower boundary is constant (30 m^2/sec) for all conditions.

It is somewhat surprising in view of the coarse treatment of the meteorological variables that the agreement between prediction and observation is so encouraging. The reasons may hinge on the predominance of the surface emission sources. During the morning hours, the level of surface concentrations is strongly controlled by the height of the inversion; the SA results show that application of the Edinger work is most suitable during this period. With these shallow mixing depths, contaminants become rather uniformly distributed up to the lid, and the surface wind is a good approximation to the transport wind for the layer. For afternoon conditions, the lid may be expected to be quite high, and subsequent inaccuracies in its computation contribute relatively little to the predicted surface concentrations. Because of the large diffusive capability of the atmosphere, errors in the winds aloft may be less significant. Equally important in the success of the model is the accuracy of the emissions field.

A Simple Analytical Model

A simple analytical method for calculating dispersion from urban area sources has been presented in a recent series of papers by researchers from the Atmospheric Turbulence and Diffusion Laboratory (ATDL) of NOAA (Gifford, 1970; Gifford and Hanna, 1970; and Hanna, 1971). The work is a welcome addition to the numerous papers on dispersion modeling, since it presents a simple method for calculating the area surface concentration that can be used to test the results of the more complex and expensive numerical models. In so doing, the authors stress an important point: "The usefulness of any . . . area source diffusion model depends on its performance compared with other area source models. In the past, as new models appeared in the literature, there was no comparison with other models. Clearly, if the concentrations predicted by a complicated model are not significantly better than the predictions of . . . simple . . . model[s] of diffusion, then there is no practical justification for the new model." The ATDL model, together with other simple formulations such as the box model or the SRI Gaussian model, can provide for such evaluations.

The ATDL model is an analytical solution of the gradient transfer form of the mass conservation equation under the following assumptions: (1) conditions are steady-state, (2) horizontal diffusion is relatively insignificant, and (3) chemical reactions and deposition are ignored. When the x-axis is aligned with the transport wind, the area concentration (χ_A) from a surface area source is determined from

$$u \frac{\partial \chi_A}{\partial x} = \frac{\partial}{\partial z}\left[K_z \frac{\partial \chi_A}{\partial z}\right] . \qquad \text{(III-12)}$$

The vertical profiles of wind and vertical eddy diffusivity are assumed to be represented by power laws, where

$$\left.\begin{aligned} u &= u_1 \ (z/z_1)^m \\ K_z &= K_1 \ (z/z_1)^n \end{aligned}\right\} . \qquad \text{(III-13)}$$

On introducing a similarity transformation of the independent variables, Equation (III-12) reduces the solution of two ordinary differential equations through the separation of variables. With the area-source distribution given over a horizontal grid (N x N), the solution at z = 0 is evaluated over N equal intervals of x:

$$(\chi_A)_o = \frac{z_1^m \ (\Delta x)^{1-s}}{c_1 u_1} \left\{ Q_1 + \sum_{i=2}^{N} Q_1 \left[i^{1-s} - (i-1)^{1-s} \right] \right\} . \qquad \text{(III-14)}$$

where $s = (m + 1)/(2 + m-n)$ and $x = N\Delta x$. If the vertical distribution of concentration is assumed to be Gaussian, the vertical standard deviation can be given by $\sigma_z = ax^b$ (see Eq. III-3) and Eq. (III-14) becomes

$$(\chi_A)_o = \sqrt{\frac{2}{\pi}} \ \frac{(\Delta x/2)^{1-b}}{u_1 a(1-b)} \left\{ Q_1 = \sum_{i=2}^{N} Q_i \left[(2i+1)^{1-b} - (2i-1)^{1-b} \right] \right\} . \qquad \text{(III-15)}$$

This simplification appears justified, since Gifford and Hanna (1970) have shown that the form of the vertical distribution of concentration has "little impact" on the surface value. However, it should be pointed out that this form of the ATDL model--Equation (III-15)--is virtually identical to the SRI Gaussian model--Equation (IV-3).

Hanna (1971) proceeds to further simplify (15) in view of the relative weight of the area source-strengths on the concentration at the receptor; if the source strength is uniform in x, the innermost area contributes 320% more to the receptor concentration than does the second sector, 420% more than the third, and so forth. Therefore, Equation (III-15) is reduced to its most simple form,

$$(\chi_A)_o = C \frac{Q_1}{u_1} \text{ , and}$$

$$C \equiv \sqrt{\frac{2}{\pi}} \left[\left(\frac{2N + 1}{2} \right) \Delta x \right]^{1-b} \frac{1}{a(1 - b)} \quad . \qquad \text{(III-16)}$$

An exponential form for Q(x) is also considered in the paper. Equation (III-16) was compared with results of more complex models as reported by Lamb (1968) for natural gas in the Los Angeles Basin and Roberts et al. (1969, 1970) for sulfur dioxide in Chicago.

Lamb's model incorporates a numerical solution to the mass conservation equation and is quite flexible, with such features as time-variable sources, space-variable wind, and ground absorption and simple chemical reactions of the pollutants. However, the model is restricted to a uniform vertical wind distribution and constant eddy diffusivities. Gifford and Hanna (1970) and Hanna (1971) compared the results of Lamb's model for a 16-hour period over 4-mile square sectors with those obtained using Equation (III-15). The results compare favorably in a qualitative sense, but no correlation statistics are presented. It is unfortunate that the ATDL computations were made using the annual average wind direction frequency distribution, together with a single value for the mean wind speed chosen on the basis of Lamb's wind speed data. It would appear that in limiting the representativeness of their data, the authors have severely restricted the value of the comparison.

The comparison with Roberts' results for Chicago is a better test of the ATDL argument. Roberts' is an integrated puff model consisting of a series of algorithms assembled around a "kernel" that represents the dispersion of contaminants from point and area sources as a three-dimensional Gaussian distribution. The kernel represents a three-dimensional puff and is integrated according to a time series of piecewise constant wind vectors and atmospheric stability parameters to "simulate the transient behavior of a continuous smoke plume."

Hanna (1971) refers to the model as "the most complex in the business." Hanna used Equation (III-16) to compute surface concentrations of SO_2 using the emissions and meteorological data given by Roberts. Hanna's predictions are about twice the magnitude of Roberts', although the former are highly correlated with observations of surface SO_2 concentration. In all cases presented by Hanna, results from the simple model provided correlations that overlapped those of Roberts within the 95% confidence interval; in a few cases, the simple model was superior.

The case presented by Hanna is not significantly conclusive to even consider the abandonment of the development of sophisticated models at this time. However, it does present a yardstick that may serve to measure the merits of the various complex models. As such, the evaluation of all models within the RAPS should be made against the output of a simple model (or models), in addition to field observations of pollutant concentrations.

Other Models

The discussion of the preceding two sections focuses on two practical approaches to the problem of simulation of atmospheric dispersion of mass (and momentum). The Gaussian models may be considered quasi-empirical, having some basis in the theory of atmospheric dynamics, while the gradient-transfer models seem to be quasi-theoretical, relying on empirical formulations for the description of wind and eddy diffusivity. The latter are appealing, since they provide a basis for examining the interrelationships or feedbacks between energetic and mass- or momentum-transfer processes. Another work that also incorporates the atmospheric feedback mechanisms is the dynamic climatology ("climatonomy") model of Lettau (1952, 1969, 1970). The work is particularly attractive in that it is centered on an analytical solution to the energy budget equations through the parameterization of the various energy transfer processes in terms of geophysical parameters; as such, feedbacks are inherent in the formulation. Lettau (1970) has applied the concept to a study of the physical and meteorological aspects and bases of urban diffusion modeling. In a practical application (1969), he investigated the nature of the radiation balance for urban areas in contrast to a rural environment.

Another type of diffusion model that should be mentioned is the application of surface-layer similarity theory. This approach has its foundation in the specification of the Eulerian characteristics of turbulent flow by the friction velocity together with the Monin-Obukhov-Lettau stability length. In contrast to K-theory, similarity theory treats the diffusion process as occurring with a finite velocity. Although physically appealing, this concept may not be of prime importance when compared with the complexities introduced by marked wind shears, irregular terrain, nonsteady conditions, and so forth. Application of similarity models seems to be further limited by the difficulties in obtaining solutions to diabatic conditions.

The statistical theory of relative atmospheric motion offers additional physical insight of the diffusion process. The original statistical theory of Taylor (1921) considered the motions of fluid particles over some diffusion time to be completely independent. Particles that thus find themselves infinitely close at some point in time must forever remain so. The relative diffusion theory provides an alternate approach in which the rate of spreading of nearby particles is considered relative to their mutual center of mass. Gifford (1968) summarizes the distinction by considering relative diffusion as the treatment of the spreading out of a cloud of fluid particles or the spreading of a plume from its instantaneous center line through joint Langrangian statistics of two dispersing particles. Single-particle Lagrangian statistics treat the average spreading of a plume about a fixed axis. The relevance of the distinction may be minimal when, for example, the objective of the computation is the prediction of hourly concentrations as in the previously discussed comment by Pasquill (1971) on the merits of the puff versus the plume formulation.

Another model based on physical principles is the vorticity-transfer formulation introduced by Lettau in 1964 and subsequently refined in a series of papers over the next five years. The theory treats the eddy displacements of fluid particles as three-dimensional, reflecting the coexisting tendencies of the fluid to conserve its initial properties while adapting to its new environment (entrainment). Additionally, the importance of the longitudinal length-scale of turbulence is considered in conjunction with the classical (Prandtl) lateral length scale. Expressions for the variances and covariances of velocity and temperature fluctuations are given in terms of the length scales and mean fluid flow properties. Non-Newtonian stress-strain (a gradient transfer) relationships result that permit eddy fluxes in nonshear regions as well as countergradient fluxes. Lettau (1964, 1966, 1967, 1968) has applied the theory to an extensive list of problems: (1) wall turbulence, (2) surface layer structure, (3) turbulent free jet, (4) Karman's similarity principle, (5) nature of eddy diffusivities for heat and momentum, and (6) heat transfer. Dabberdt (1969) has extended the theory to the case of bidirectional mean shear flow in the planetary boundary layer. The theory may provide a rational basis for prescribing the nature of the horizontal eddy diffusivity.

Moses (1969) discusses a tabulation prediction scheme developed at the Argonne National Laboratory. The method consists of developing an ordered set of combinations of meteorological variables and presenting the distribution of surface concentrations of a particular contaminant for each element in the set. In the tabulation, the independent variables are wind direction and speed, time of day, temperature, and stability; various percentile values are presented in addition to maxima and minima. The procedure is purported to have the following advantages over other receptor-oriented methods: (1) ease of use, (2) speed, (3) presentation of percentile distribution of concentration (forecast application), and (4) implicit consideration of nonlinear relationships of the independent variables.

The disadvantages of the method are: (1) an extensive observational data base (reported to be a 2-year minimum) is required at each forecast (receptor) point, (2) a large digital computer is required for the initial tabulation, (3) changes in the emissions grid degrade the results, and (4) the model cannot account for aerodynamic effects of construction. However, the method appears attractive for regions where continuous records of meteorological, emissions, and air quality data are available. It would seem to have application in the quantification of a normalized prediction of the spatial distribution of contaminants from analytical or numerical models.

Finally, a very recent multibox model developed by MacCracken et al., (1972) at the University of California should also be mentioned. The model is simple and flexible. First, observed time- and space-dependent values of surface wind and inversion height are put into a meteorological data model that incorporates topographic effects and develops a mass-consistent horizontal wind field and diffusion coefficients (vertical and horizontal) for the region. These are then used together with a time- and space-varient emissions field in the multibox model to compute time-varying pollutant concentrations in each box. Box-size resolution is variable and photochemical reactions are incorporated in the model. Preliminary verification tests have been conducted for the simulation of CO concentrations in the San Francisco Bay Area with some encouraging results.

Additional work will concentrate on refinements to the photochemical aspects, as well as on an evaluation of the sensitivity of the model to input data representation.

The models and types presented are not intended to provide an exhaustive survey of air quality simulation methods. Rather, the intent has been to supplement the two basic types--Gaussian and gradient-transfer--with representative alternatives that possibly have less general application to the overall modeling problem but that can provide varying degrees of insight or utility.

Numerical Weather Prediction Models

The use of currently available, numerical weather prediction models should be considered to provide predictions of the meteorological variables for use as input data to the regional atmospheric dispersion models.

Their application to air pollution problems can make use of the tremendous advances that have been achieved in numerical weather forecasting in the last 15 years. On the other hand, special problems exist in making detailed forecasts for areas of limited size. These problems are discussed briefly below.

The numerical models that are in operational use for weather prediction encompass either the Northern Hemisphere or the entire globe. In the United States, such models are in use at the National Meteorological Center, NOAA; at the Air Force Global Weather Central, Offutt Air Force Base, Omaha; and at the Navy's Fleet Numerical Weather Facility, Monterey, California. These models represent the atmosphere using 6 to 10 levels in the vertical and a horizontal mesh with points separated by 300 to 400 km. The atmospheric boundary layer structure and dynamics are treated in only a rough and approximate manner. These models are used primarily for synoptic scale forecasting for periods up to three days in advance.

To obtain more detailed forecasts, the Air Force uses a mesoscale model (Kerlin, 1970) for North America, which is similar to the hemispheric model except that the horizontal grid spacing is only half as large. This horizontal spacing (~150 km) is considerably smaller than the average distance between radiosonde stations, so that further reduction would not be beneficial, unless additional radiosonde observations became available. However, it is possible to obtain additional vertical resolution within the atmospheric boundary layer from radiosonde data. This is done in the Air Force boundary layer model (Hadeen, 1970) which uses the same horizontal grid as the mesoscale model for an area covering the contiguous 48 states. The boundary layer model has seven layers (or eight levels) between the surface and 1600 meters and provides forecasts of winds, temperature, moisture, and pressure for periods of 12 to 24 hours in advance. This is made possible because the boundary layer model is controlled to a considerable degree by information at its upper level (about 1600 m aboveground) produced by the mesoscale forecast model mentioned above. The winds and pressure at this level predicted by the mesoscale model control the forecast positions of features such as highs, lows, and fronts in the boundary layer.

The mesoscale model also provides forecasts of clouds for layers above 1600 m. The boundary layer model has internal dynamics that produce temperature forecasts based on advection, diurnal heating effects, and cloudiness. These temperatures give thermal winds that are used to determine surface winds from the upper level values provided by the mesoscale model. The intervening winds are then fit to a Blackadar type of Ekman spiral. The model follows work originally done by Gerrity (1967). The equations also include terms for eddy diffusivity and conductivity, the effect of cloudiness on diurnal heating, and changes of state of water substance. In spite of certain simplifications and assumptions, the Air Force boundary layer model is by far the most complete model for low level meteorology that is now available on an operational basis. The information from the model is used in forecasting ceilings and visibility at airports, low-level turbulence, severe thunderstorm occurrences, and so forth.

It is strongly recommended that EPA consider obtaining forecasts from this model for the four (or more) grid points that are nearest to St. Louis. It is our understanding that the Air Force Global Weather Central is willing to make the forecasts available to other government agencies. Similar boundary layer forecasts are not expected to become available from NMC for two years or more. One of the major research tasks proposed for the Regional Study concerns the use of these boundary layer forecasts in controlling air quality within a region. This will be an entirely new line of research.

Further development of even more detailed boundary layer numerical models most logically would introduce much finer horizontal grids (~10 km instead of the ~150 km of the present Air Force model), more vertical levels, and more complete and complex dynamics. Work in this direction has been done by Pandolfo (1965 and 1971). In addition to purely scientific problems, some serious practical obstacles would be encountered in applying such a model for operational purposes. These entail the difficulty of obtaining initial conditions for the entire mesh and also the necessity of specifying upper and lateral mathematical boundary conditions as the prediction proceeds. This can be understood from the considerations discussed below.

To obtain detailed initial conditions for a fine-mesh regional model covering an area of 100 km on a side would be quite feasible using a dense network of observations. However, under moderate or strong winds, air will cross such an area within a few hours. After the original air has passed out of the region, the conditions predicted within the region are controlled to a large degree by assumptions made concerning the time history of meteorological properties at the inflow boundaries. This tends to limit the predictability of the model to a few hours, unless accurately predicted conditions at the upper and lateral boundaries of the fine mesh are available from some other source, such as the mesoscale numerical forecast.

At this time we do not envision a realistic method for obtaining such detailed lateral conditions, since the desired information is beyond the resolution of the mesoscale model. The natural alternative is to expand the area of the fine-mesh model and of the observations needed for its initial conditions. However, to encompass air that could reach the area of interest within a longer period such as 12 hours, one would have to expand the dimension of a side of the fine mesh to at least 500 km. Obtaining detailed data over a region this large would probably be prohibitively expensive. Thus, it does not seem likely that operational forecasts for a finer mesh than that used by the Air Force can be achieved within the next few years for this reason alone.

Another anticipated difficulty with fine-mesh boundary layer predictions is that the finite difference methods in current use have the general property of tending to smooth the smaller scale features of the weather patterns as the prediction proceeds. Therefore, wanted details tend to be lost as the period of prediction increases.

A final point about fine-scale predictions for a region concerns the computational requirements. These depend on the number of grid points in the mesh, the number of variables carried, the efficiency of iteration schemes for relaxation or similar processes, and the length of the time step. It is instructive to compare a possible regional model to a known global model. For example, Manabe et al. (1971) used a horizontal mesh of 9200 points (~250 km spacing) at 9 levels, giving approximately 80,000 points for the globe. We assume that that their dynamical equations and those of a fine-mesh regional model are of similar nature and complexity. Their time step was 5 minutes. Obtaining a 24-hour global prediction required 27 hours of computation using a Univac 1108 computer. A regional fine mesh model of, say, 100 horizontal points (~10 km spacing) at 10 levels would give only 1000 points. However, the time step for computational stability would have to be reduced to 5 (10/250) or

0.2 minutes because of the smaller mesh size. Thus, the computational requirements for a fine-mesh regional model are about (1000/80,000) (5/0.2) = 0.3 times as large as those required by Manabe. These requirements are still considerable and would entail large computer costs and a sizable staff to man the operation.

The difficulties and costs enumerated above would have to be surmounted to obtain a fine-scale regional prediction of boundary layer meteorological conditions. A continuing research program should be directed to this subject. However, we believe that for the next few years main reliance should be placed on forecasts provided by the present Air Force boundary layer model.

Model Sensitivity to Meteorological Variables

It is particularly important to assess the sensitivity of the various models to variations in the input meteorological variables; evaluation of the sensitivity to emission and reaction factors is equally important, but discussion will be deferred to subsequent sections in keeping with our modular approach to modeling theory. The basic objective of evaluating the sensitivity of model calculations is to assess the importance of the various atmospheric processes in determining concentration levels and to simultaneously permit the refinement of model formulations based on suitable observations of the relevant parameters. The interchange between observations and simulation is reciprocal, and it will be particularly important that the observing network not retard simulation development. However, in designing the field system, it will be necessary to be guided by available modeling results, as well as by the experience of past observation programs.

Specifically, it is important to know the effects in output values caused by changes in the inputs. If a very small change in a certain input results in a large change in the output, then the model--and presumably the physical system--is very sensitive to that input and it will be necessary to provide values of the input that are very accurate to avoid large errors in the output. On the other hand, if large changes in an input have little effect on the output, then little effort need be expended to achieve high input accuracy for that particular variable. Sensitivity is not likely to remain constant over the whole range of values that a particular variable can assume. Thus it is possible that for certain combinations of parametric values, the model output will be totally independent of the changes in one or more inputs.

independent of the changes in one or more inputs.

Of the dispersion models discussed in the preceding sections, the gradient-transfer types have the greatest potential for describing the atmospheric dynamics and energetics on the regional scale. The extensive and complex numerical solutions are adaptable to synthesizing the intricacies of the four-dimensional meteorological fields, but their sensitivity can be evaluated only through a systematic variation of the inputs (either singularly or in combination). The process is tedious and expensive, and no systematic evaluations have been reported to date. Several limited tests have been made (see, for example, Roth et al., 1971, and Pandolfo et al., 1971), but the results are given only in qualitative terms.

Gifford and Hanna (1971) have evaluated the response of the predicted concentration from their K-model to normalized changes of the input parameters (P),

$$\frac{\delta(\chi_A)_o}{(\chi_A)_o} = k \frac{\delta P}{P} \quad , \qquad \text{(III-17)}$$

using an alternate form of Equation (III-14):

$$(\chi_A)_o = \frac{z_1^m (\Delta X/2)^{1-s}}{c_1 u_1 \;(B(1 - s)} \left\{ Q_o + \sum_{i=1}^{N} Q_i \left[(2i + 1)^{1-s} - (2i - 1)^{1-s} \right] \right\} \; , \qquad \text{(III-18)}$$

Where the parameter B depends on the shape of vertical concentration profile. The results of the analysis are given in Table III-1. The model is shown to be fairly sensitive to variations of the stability parameter (1-b) under stable conditions; otherwise, small changes in the input parameter produce only small changes in the surface concentration.

Table III-1

SENSITIVITY ANALYSIS OF THE GIFFORD AND HANNA (1971) DISPERSION MODEL

Parameter	Range	$\delta \ell n(\chi_{A})_{o} = k \delta \ell n P$	k
u	1-30m sec^{-1}	$- \delta u/u$	-1
Δx	5-50 km	$\frac{\delta \Delta x}{\Delta x}(1-b)$	(1-b)
1-b	.09 - .29	$\frac{\delta(1-b)}{(1-b)}(1-b)x^{1-b}\ell n x$ (for $Q_1 = Q_o$)	$(1-b)x^{1-b}\ell n\, x$
N	5 - 10	$\frac{\delta(N+1)}{(2N+1)}(1-b)$	(1-b)
Q_o (Central source box)	Many orders	$\frac{\delta Q_o}{Q_o}\left(1 + \sum_{1}^{N} \frac{Q_i}{Q_o} F_i\right)^{-1}$	$\simeq$0.6 for type D and $Q_i = Q_o$
Q_j (j'th source box)	Many orders	$\delta Q_j F_j \left(\frac{Q_o}{Q_j} + \sum_{i=1}^{N} \frac{Q_i}{Q_j} F_i\right)^{-1}$	$\simeq$.10 to .15 for type D and $Q_i = Q_o$
B	0.4 - 1	$- \delta B/B$	-1
f_i	.01 - 1	$\delta f_i/f_i$	1

In the above, $F_i = (2i + 1)^{1-b} - (2i - 1)^{1-b}$. The result for (1 - b) was obtained, assuming for simplicity that $Q_i = Q_o$ = constant, from the continuous form of Equation (III-18).

$$\chi_{AO} = (2/\pi)^{1/2} Q_o x^{1-b} [a(1-b)u]^{-1} \quad .$$

In view of the small variability of a(1-b) over the expected range, from unstable to type-D conditions (neutral stability), this product was assumed constant in evaluating the effect of (1-b).

On the basis of this analysis, Gifford and Hanna conclude that surface concentrations resulting from area-source emissions are relatively insensitive to the form of the vertical concentration distribution (B), and they "are not inclined to regard uncertainty about the precise form of this quantity as being much of a problem." This hypothesis should be tested with other models that incorporate vertical profiles of diffusivity that are nonmonotonical and wind that is bidirectional, as well as the important case of elevated sources.

Michael (1971) has evaluated the effects of the shape of the boundary layer wind and diffusivity profiles on the center-line distribution of surface concentration from an elevated point source. The form of the mass conservation equation used is:

$$u \frac{\partial \chi}{\partial x} + v \frac{\partial \chi}{\partial y} = K_y \frac{\partial^2 \chi}{\partial y^2} + \frac{\partial}{\partial z} \left(K_z \frac{\partial \chi}{\partial z} \right) \quad . \qquad \text{(III-19)}$$

The wind and diffusivity profiles were not determined uniquely but are assumed to have the shapes illustrated in Figure III-1. The lateral diffusivity was assumed to be independent of height. The results of various combinations of diffusivity and unidirectional wind profiles are given in Figure III-2; the source height h was taken to be 100 m. The important point shown by these graphs is that fall-off of the peak surface concentration at large distances from the source is determined by the shape of the profile of diffusivity and not velocity (however, it must be kept in mind that the diffusivity profile is controlled in part by the velocity profile). The velocity profile, in turn, affects the location and magnitude of the maximum and close-in distribution.

The effect of directional shear on the ground-level concentration is illustrated in Figure III-3. The downwind distribution of relative concentration is controlled by the shear to a significant extent at long travel distances but minimally near the source.

On the other hand, Gaussian models have been rather extensively evaluated with regard to their sensitivity to parameter variations. A thorough treatment of the problem has been given by Hilst (1970), using the Travelers Research Corporation (TRC) regional model where the effects of both random and systematic input errors were examined. The sources of error and uncertainty were grouped into four classes: (1) pollutant diffusion and distribution estimates, (2) positional errors, (3) source strength estimates and (4) pollutant decay and loss estimates. The specific sources of errors that constitute the four classes are shown later in Figure III-4; taken from Hilst.

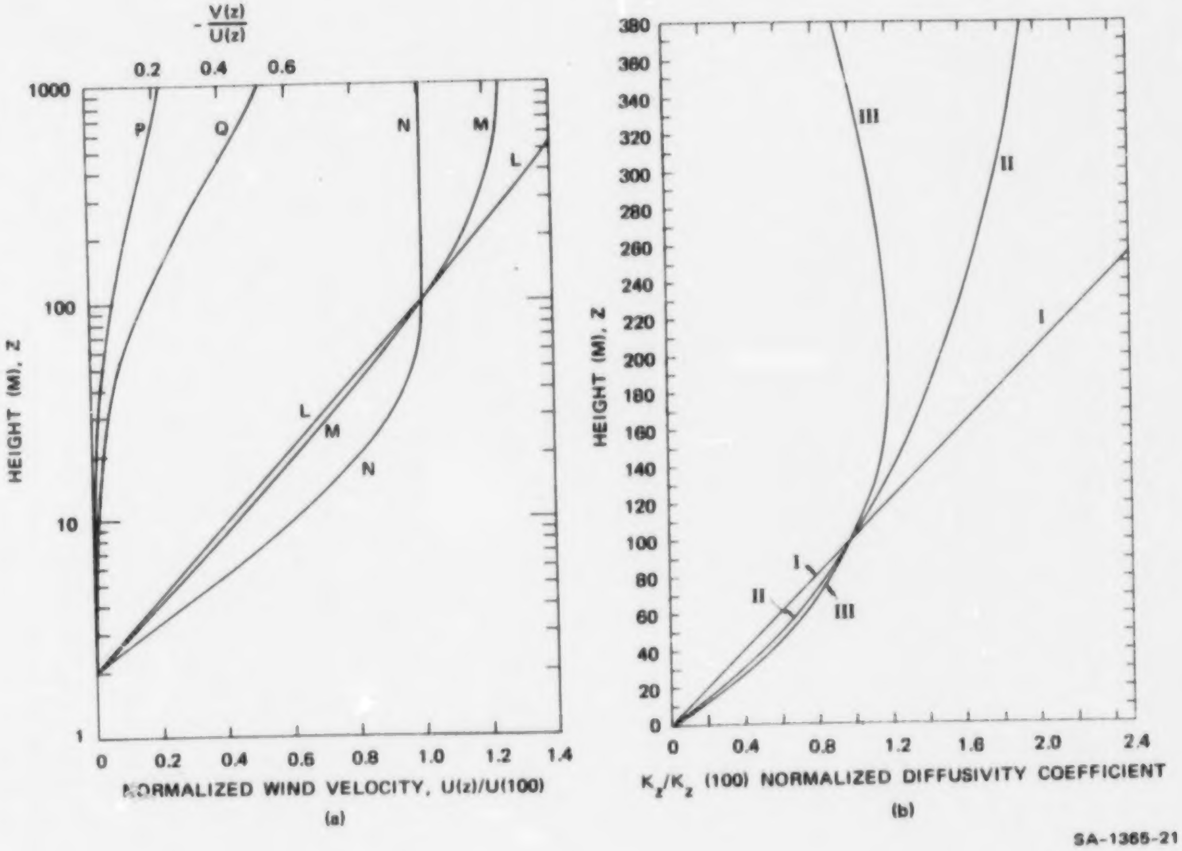
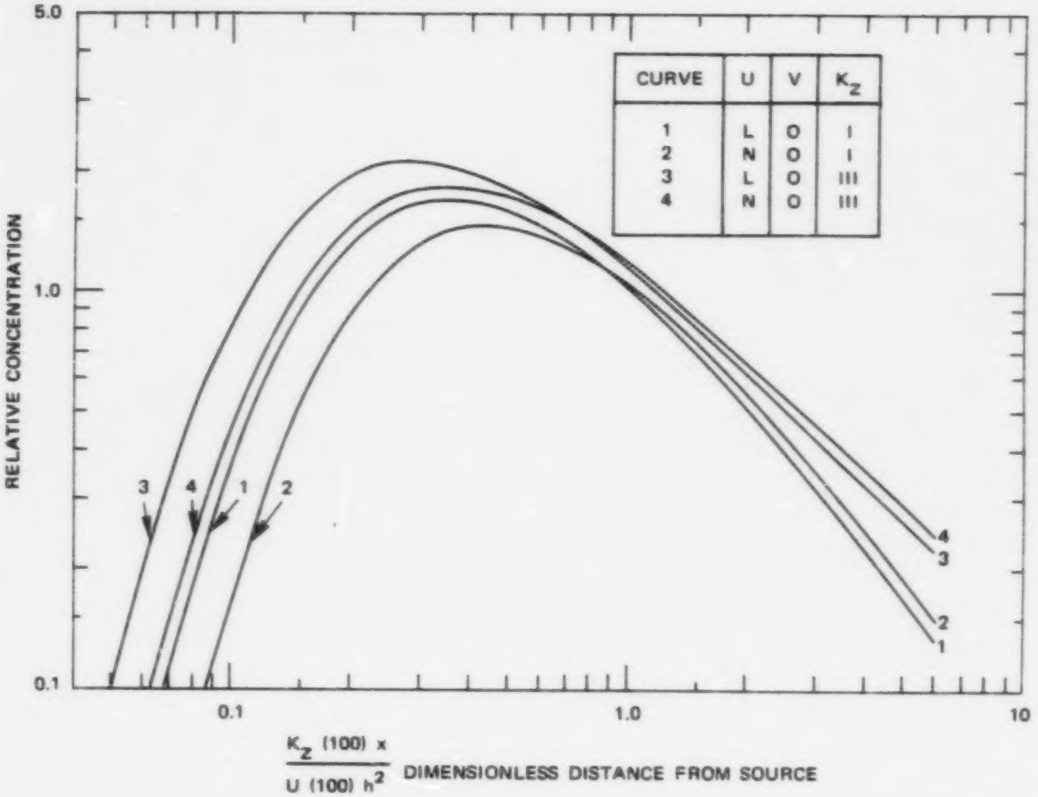


FIGURE III-1 VERTICAL PROFILES OF WIND AND EDDY DIFFUSIVITY (MICHAEL, 1971)



CURVE	U	V	K_Z
1	L	O	I
2	N	O	I
3	L	O	III
4	N	O	III

SA-1366-22

FIGURE III-2 PEAK SURFACE CONCENTRATION AS A FUNCTION OF DISTANCE FROM AN ELEVATED SOURCE (MICHAEL, 1971)

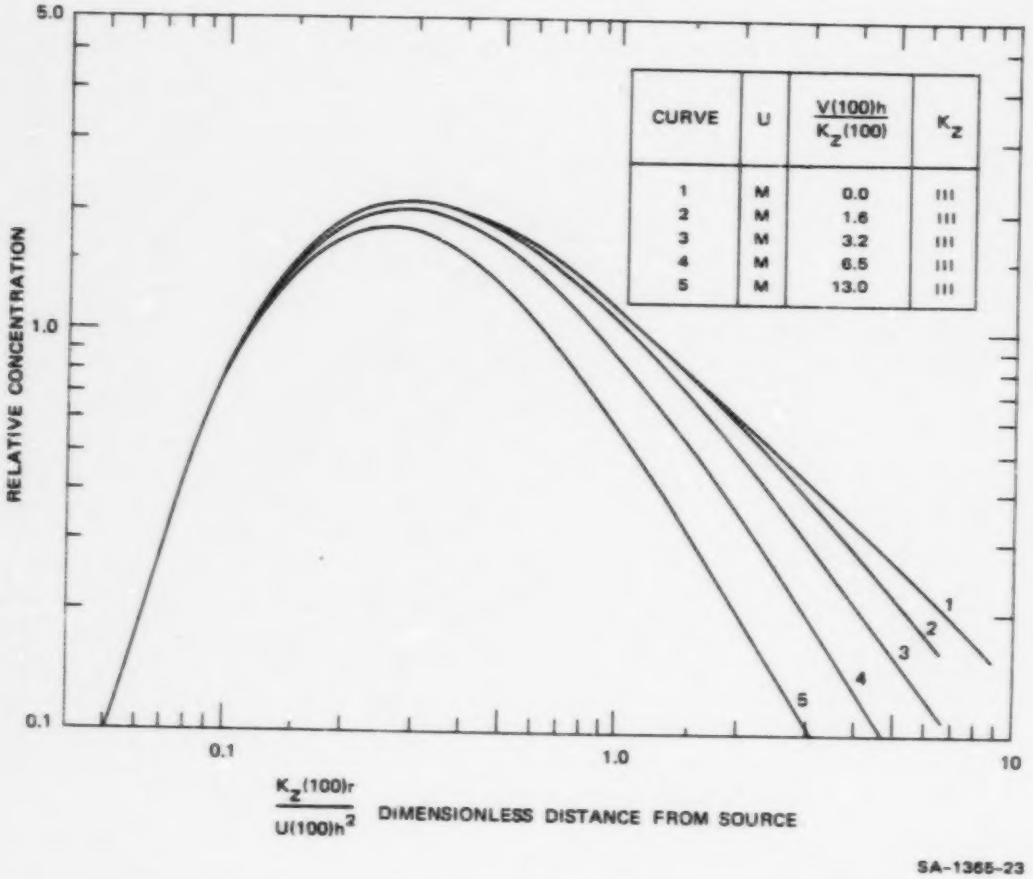


FIGURE III-3 PEAK SURFACE CONCENTRATION AS A FUNCTION OF THE RADIAL DISTANCE FROM THE SOURCE (MICHAEL, 1971)

The distinction between the effects of random and systematic errors is particularly interesting. For example, random errors are to be expected in the preparation of source inventories, but these are likely to be complemented in some cases by a systematic underestimate through the neglect of singularly unimportant--yet collectively significant--sources. Errors in diffusion estimates will tend to be primarily random for short averaging times but systematic for longer periods. Positional errors can also be of both types; surveying errors will usually be random, but the result on the positioning of sources or receptors with respect to a given trajectory may be systematic (especially for small source-receptor separation). Results of Hilst's sensitivity analyses for the meteorological parameters follow, with only the systematic errors actually evaluated. The effects of systematic and random errors in the source field are given in the emissions section.

The TRC model uses a receptor-oriented Gaussian plume formulation. Contributions to the concentration at a receptor are made from sources along the air trajectory (not necessarily straight); wind and dispersion fields are treated as steady over two-hour periods. The test region was the State of Connecticut.

The effect of the lateral diffusion coefficient on the concentration resulting from a multiple-source field is minimal, because the effect of lateral diffusion from a distributed source field is to rapidly produce a uniform crosswind concentration. This is reflected in Table III-2 where values of the mean and standard deviation of the normalized difference between predictions of concentration with and without a systematic lateral diffusion error are given as a function of the latter.

Sensitivity of the predicted concentration to systematic errors in the vertical diffusion coefficient is significant, as shown in Table III-3. This is particularly true when the coefficient is underestimated $[(\sigma_{z-T} - \sigma_{z-E})/\sigma_{z-T} \rightarrow 1]$. The effect is less significant for overestimates partially because of the predominance of the effects of local sources.

The effects of positional errors (e.g., mislocation of sources, receptors, and trajectories) affect the nature of the errors of the other parameters. Hilst used systematic variations of the wind direction for the assessment of the interrelated, positional errors. The mean errors are comparable with those associated with the vertical diffusion coefficient, while the variances are even larger as shown in Table III-4.

Table III-2

CALCULATED VALUES OF THE MEAN AND STANDARD DEVIATION OF $[(\chi_T - \chi_E)/\chi_T]$ * AS A FUNCTION OF SYSTEMATIC ERRORS IN THE LATERAL DIFFUSION COEFFICIENT σ_Y, AFTER HILST (1970)

$\left(\frac{\sigma_{Y-T} - \sigma_{Y-E}}{\sigma_{Y-T}}\right)$	$\overline{\left(\frac{\chi_T - \chi_E}{\chi_T}\right)}$	$\sigma_{\chi(T,E)}$
-0.2	-0.003 ±0.02†	0.105 < 0.122 < 0.139†
+0.2	0.007 ±0.03	0.130 < 0.151 < 0.172
-0.5	-0.018 ±0.03	0.138 < 0.161 < 0.183
+0.5	0.012 ±0.03	0.138 < 0.161 < 0.183
-0.7	-0.032 ±0.04	0.170 < 0.197 < 0.224
+0.7	0.014 ±0.03	0.141 < 0.164 < 0.187

* $\chi_T \equiv$ "true" concentration (no error)

$\chi_E \equiv$ "estimated" concentration (with error)

† Limits are assessed at the 95% confidence level.

Table III-3

CALCULATED VALUES OF MEAN AND STANDARD DEVIATION OF $(\chi_T - \chi_E)/\chi_T$ AS FUNCTION OF SYSTEMATIC ERRORS IN VERTICAL DIFFUSION COEFFICIENT σ_z, AFTER HILST (1970)

(Limits Assessed at 95% Confidence Level)

$\left(\frac{\sigma_{zT} - \sigma_{zE}}{\sigma_{zT}}\right)$	$\overline{\left(\frac{\chi_T - \chi_E}{\chi_T}\right)}$	$\sigma_{\chi(T,E)}$
-0.2	0.055 ± 0.03	$0.112 < 0.130 < 0.148$
+0.2	-0.071 ± 0.03	$0.143 < 0.164 < 0.187$
-0.5	0.120 ± 0.03	$0.133 < 0.155 < 0.177$
+0.5	-0.236 ± 0.05	$0.206 < 0.240 < 0.276$
-0.7	0.151 ± 0.03	$0.145 < 0.168 < 0.191$
+0.7	-0.429 ± 0.15	$0.669 < 0.776 < 0.885$

Table III-4

CALCULATED VALUES OF MEAN AND STANDARD DEVIATION OF $(\chi_T - \chi_E)/\chi_T$ AS FUNCTION OF SYSTEMATIC ERRORS IN WIND DIRECTION (θ); HILST (1970)

(Limits Assessed at 95% Confidence Level)

$\theta_T - \theta_E$, degrees	$\overline{\left(\frac{\chi_T - \chi_E}{\chi_T}\right)}$	$\sigma_{\chi(T,E)}$
+10	-0.134 ± 0.04	$0.191 < 0.222 < 0.253$
-10	-0.124 ± 0.04	$0.193 < 0.218 < 0.248$
+20	-0.436 ± ?	-
-20	-0.232 ± 0.26	$1.14 < 1.32 < 1.51$
+40	-0.152 ± 0.22	$0.93 < 1.08 < 1.23$
-40	-0.524 ± 0.33	$1.45 < 1.68 < 1.92$

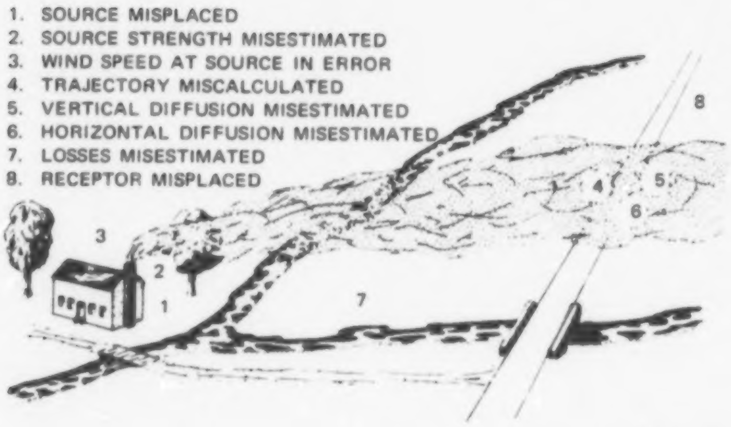
As a result of the TRC study, the sensitivity of the calculations to variations of the input meteorological factors may be summarized by considering the mean and standard deviations of the prediction error resulting from normalized variations of ±0.5 of the input parameters (± 20, 40° for wind direction):

Type of Error	Mean Error	Individual Errors
Wind direction (± 20°)	≤ ~ -25%	-200 to +150%
Wind direction (± 40°)	≤ -50%	-275 to +175%
Vertical diffusion coefficient	≤ ± 25%	≤ ± 50%
Lateral diffusion coefficient	≤ ± 5%	≤ ± 20%

It should be recognized that while the TRC work is extensive, it is still a case study for a given emissions network and meteorological field. As such, these results generally may not be applicable, yet they do illustrate the manner in which the concentrations can respond to variations of the appropriate pollution variables within a given model design. However, the model does not explicitly consider the effects of a limited mixing depth.

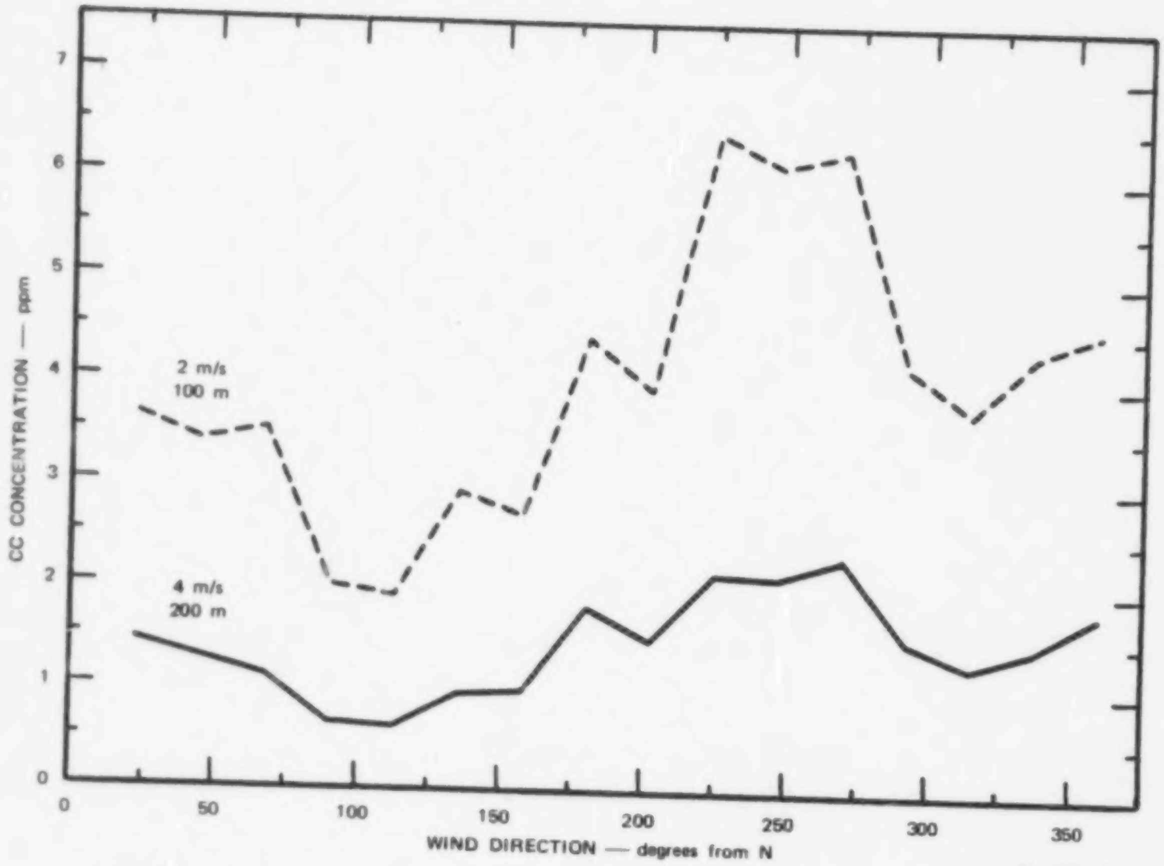
The SRI dispersion model has also been evaluated (Ludwig et al., 1970) for its sensitivity to variations in the meteorological and positional factors (as well as in the emissions field) in terms of: (1) wind speed, (2) wind direction, (3) stability and mixing depth, and (4) receptor location. The first-order response of predicted concentrations to wind speed changes by the SRI model is directly proportional to the inverse of that parameter. This is the case with both the general Gaussian formulation and the specific box model used with limiting mixing depths. A second-order response occurs in the effect of wind speed on the determination of the stability category and is considered in terms of sensitivity to variations of the latter.

The ramifications of wind direction errors are illustrated in Figure III-4 and are seen to be closely related to sensitivity with regard to the source field. Figure III-5 is an example of the changes that can result and is based on sample calculations for the Washington, D.C., CAMP station under two sets of meteorological conditions. Calculations were made at 22.5° increments, and the largest difference between adjacent directions is about 60%. The effects of receptor location are


1. SOURCE MISPLACED
2. SOURCE STRENGTH MISESTIMATED
3. WIND SPEED AT SOURCE IN ERROR
4. TRAJECTORY MISCALCULATED
5. VERTICAL DIFFUSION MISESTIMATED
6. HORIZONTAL DIFFUSION MISESTIMATED
7. LOSSES MISESTIMATED
8. RECEPTOR MISPLACED
1
2
3
4
5
6
7
8

SA-1365-24

FIGURE III-4 SOURCES OF INPUT ERRORS AND UNCERTAINTIES IN VARIABLES UTILIZED IN AIR QUALITY SIMULATION MODELS (HILST, 1970).



TA-7874-6R2

FIGURE III-5 EFFECT OF WIND DIRECTION ON CONCENTRATIONS COMPUTED FOR THE WASHINGTON, D.C., CAMP STATION. (0700-0800 LST, Neutral Stability, Wind Speeds and Mixing Depths as Shown).

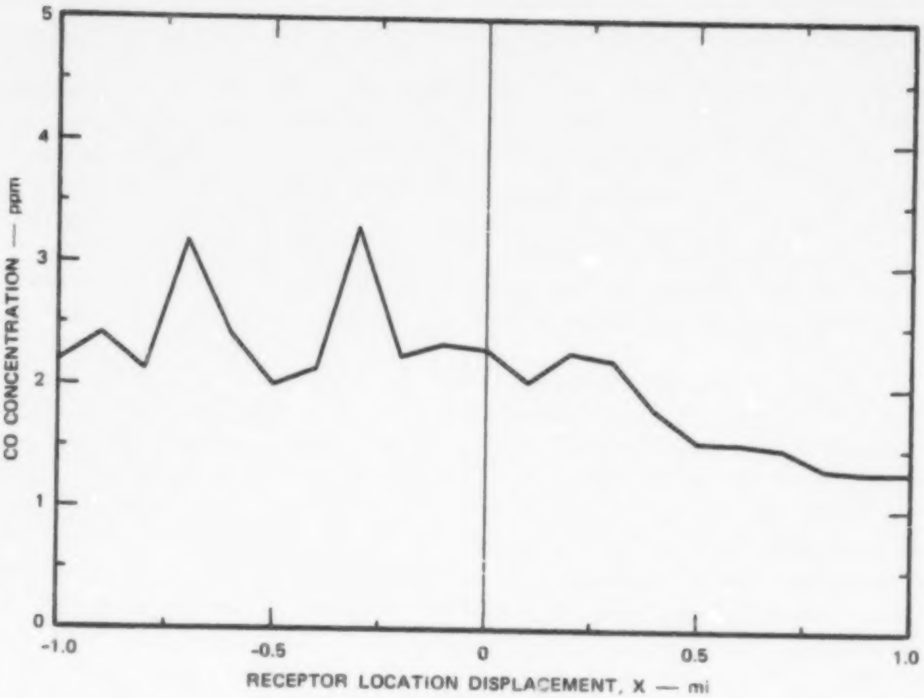
closely related to those of wind direction, i.e., moving the receptor relative to the source alters the relative location of the sources (or the wind direction). To study the effects, the receptor location was displaced by 0.1 mile increments; Figures III-6(a) and 6(b) show the changes in calculated concentrations arising from both lateral and longitudinal displacements. The initial location again corresponds to the CAMP station. In this area, the street network is dense, and concentrations are quite sensitive to the location of the receptor relative to the streets.

Stability and mixing depth interact in the SRI model to determine which of the two submodels (Gaussian or box model) is used for the calculation of the contribution of emissions from a given upwind segment. To determine the effects on the model output of variations in stability and mixing depth, calculations were made using two different configurations of sources. In the first case, source strengths were assumed to be the same in all segments. It was assumed that there were no emissions beyond the outer boundary of the last segment, 32 km from the receptor. The results of calculations for this source configuration and for various combinations of stability and mixing depth are shown in Figure III-7(a). The results are essentially the same for low values of mixing depth, regardless of stability. For very shallow mixing layers, the box model is used for almost the entire distance upwind of the receptor. Since the box model results are independent of stability, it is to be expected that the results converge at small mixing depths.

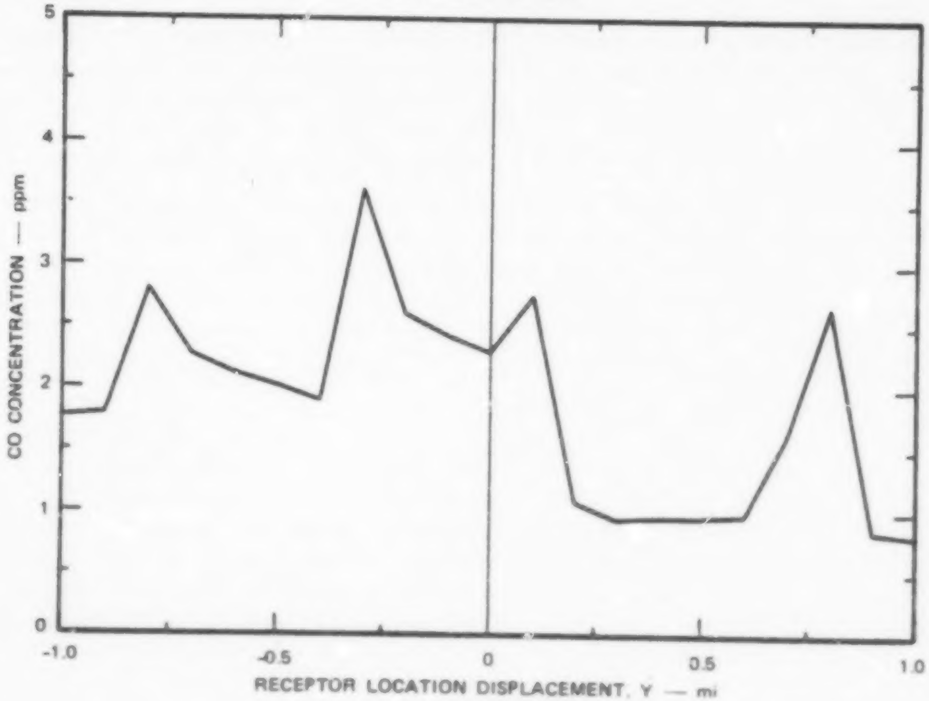
The neutral and slightly unstable cases are unaffected by mixing depth for the larger values of the parameter. This also is quite reasonable, because the mixing of the emitted material proceeds rather slowly under the more nearly stable conditions and the mixing is uninhibited by the top of the mixing layer, if it is sufficiently high. In such cases, the Gaussian model is used for the concentration calculations throughout the entire 32-km upwind distance. The results of Gaussian model calculations are independent of mixing depth.

For the moderately unstable and extremely unstable cases, the box model is used in the calculations, for at least some segments, for mixing depths to 3000 m. However, the number of segments for which the box model is applicable decreases with increasing mixing depth, so that calculated concentrations become less dependent on mixing depth as that parameter increases. This is reflected in the continual approach of the slope of the curves toward zero.

The SRI sensitivity analysis for mixing depth and stability was recalculated with a source strength that decreased linearly from the receptor to zero 32-km upwind. The results of these calculations are shown



(a) **ALONG-WIND**



(b) **CROSS-WIND**

TA-7574-5R

FIGURE III-6 EFFECT OF MOVING RECEPTOR POINT (Washington, D.C., CAMP Station, 0700-0800 LST, Wind 270°/4 ms^{-1}, Neutral Stability, Mixing Depth 200 m)

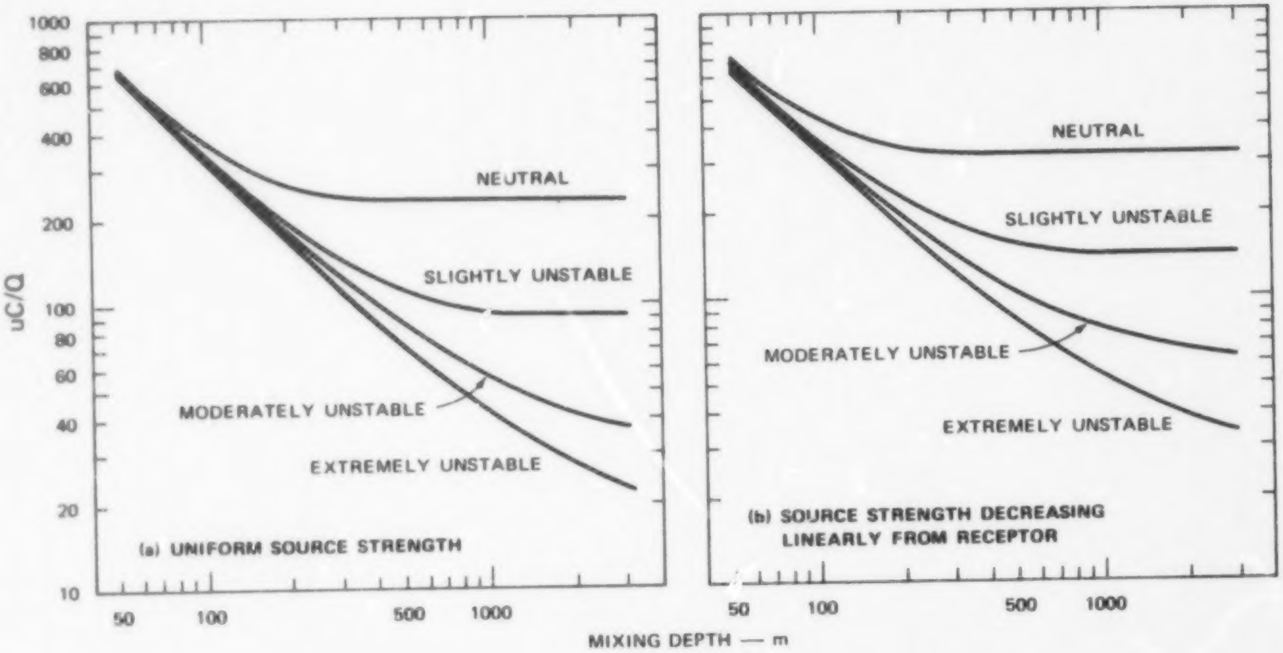


FIGURE III-7 NORMALIZED CONCENTRATION AS A FUNCTION OF STABILITY AND MIXING DEPTH. Normalized for average source strength over 32-km upwind sector.

in Figure III-7(b). Qualitatively, the two source configurations produce similar results. However, the dependence on mixing depth is less for all cases for the latter.

Evaluation of model sensitivity is clearly a difficult task. The models discussed exemplify the complexities that arise as a result of the method of solution of the equations, as well as of the representation of the variables. Further complicating the problem are the discontinuities that exist, for example, in given emissions and topographic (urban-rural differences) fields, as well as in the variations between regions. The effects of relatively small-scale horizontal wind variations (< 5 km) must also be defined.

Experimental Meteorology Program

General

The experimental meteorology program will cover field experiments that are directed toward understanding and subsequently describing the relationships between atmospheric dynamic, kinematic, and energetic processes and the resultant transport and diffusion of effluents in the lower atmosphere. A basic concept behind these experimental studies in RAPS is that they will be conducted both over an extended period of time and intensively during specified periods; thus the data will serve to show not only the nature of atmospheric processes for specific periods but also the range of variability of conditions that can be expected. This latter feature is extremely important, because in many air pollution situations not only the long term average contributes to the problem but also the infrequent, short term departures. The RAPS program will be especially designed to document the frequency and magnitude of these situations.

Tracer studies anticipated for the RAPS program for the study of transport and diffusion processes will use both gaseous and particle tracers in addition to neutral-density balloons. Techniques that include a significant amount of automatic operation will fit best into this program, because they can be used more readily on a semiroutine basis for extended periods with minimum manpower. For gaseous tracer studies, tracers such as sulfur hexafluoride (SF_6) are preferred, because the analysis is automated more readily and can be undertaken at a field laboratory soon after the test. This is not the case with insoluble fluorescent pigments, which generally require visual counting using a microscope. Soluble pigments and dyes have some uses as tracers and can be analyzed rapidly in the field. Additional gaseous tracers should be developed and tested to augment SF_6 because of increasing background levels of SF_6 and to permit dual or multiple point releases as discussed later.

The RAPS experimental meteorology program should include a facility for the mapping of horizontal wind trajectories using constant-level balloons at one or more altitudes. The operational facility should permit target balloons to be followed for distances of at least 150 km at levels as low as 150 m. Two systems, in particular, are potentially available for these experiments: (1) conventional radar tracking and (2) a new radio-locating system using Doppler principles.

The radar system has been developed to a high degree of sophistication, principally by the Atmospheric Sciences Laboratory at NOAA, and its advantages and limitations are fairly well known. The system uses a van-mounted M-33 radar to track one or more transponder-equipped balloons. Radar position data are recorded on magnetic tape and subsequently processed by computer. NOAA has tested and refined the technique in a series of field programs over the past decade. Limitations are in the operating costs of the system and the current inability to provide real-time, quantitative wind data.

The radio-locating system has been designed and given preliminary field testing as a proprietary system by Control Data Corporation (Belmont, 1971) under the trade name METRAC. The CDC system provides position location using Doppler radio techniques and a multistation network, together with transponder-equipped balloons. The data are transmitted to a central computer for recording or real-time processing and display. METRAC is a flexible and attractive system, and the feasibility of incorporating it or an equivalent system as an integral part of the Regional Study is strongly encouraged; operational details follow in a subsequent discussion.

Additional experimental meteorology programs and studies will also complement and supplement the basic research network. As discussed later, these include radiation studies, supplemental aerial sampling, and potential satellite applications.

Tracer Studies of Transport and Diffusion

Purpose and Scope

An obvious goal of the overall field program is the development of the capability to use operational meteorological observations for the assessment of the dilution capabilities of the atmosphere over an urban and adjacent rural area. As work progresses toward this goal, a system for the routine collection of essential meteorological data will be established for estimating pollutant transport and diffusion and eventually for pollution forecasting. Such a system, while retaining several special observations at key points, might include deployment of standard instrumentation over a subsynoptic scale no larger than one-half degree of latitude.

Over areas free of unusual irregularities in terrain, it is assumed that with the observational data it is possible to describe relevant atmospheric characteristics that also account for the transport and dispersion of atmospheric pollutants. To test the validity of this assumption so as to check numerical models and to aid in the design of an optimum system of meteorological observation, it is considered necessary to conduct field studies to directly observe atmospheric dispersion. Tracer studies are one technique in which known amounts of identifiable materials (gases and particles) are released and sampled downstream at other positions and times. The release of balloons that float at a constant altitude and thus serve to track the transport wind is another technique for tracing. The introduction of tracers enables the analyst to start with a known source at a specific location and to avoid confusion with other sources in subsequent measurements downstream. During the proposed tracer studies for the RAPS program, measures of the tracer dispersion (in terms of standard deviations of integrated concentrations) and its variations would be related to measures of atmospheric turbulence and transport; atmospheric indices in turn would be related to meteorological analyses based on observations that are feasible for routine acquisition.

A good review of the meteorological aspects of diffusion and of experimental tracer studies is given by Slade (1968); other good reviews covering studies into the late 1960s are also available (Stern, 1968, and WMO, 1970). Past studies have provided the background information required for the RAPS program. In fact, the St. Louis Dispersion Study of McElroy and Pooler (1968) during 1963-65 may serve as a prototype tracer study. Other tracer work was carried out in the early 1950s by Leighton and Ditmar (1952). With the exception of the programs reported by Hilst and Bowne (1966), and the tetroon experiments of Pack and

Angel (1963), most of the tracer experiments have not dealt directly with the urban environment. Many of the previous field studies, as a result of economic limitations, were conducted without sufficiently detailed meteorological measurements and without adequate modeling of existing and changing atmospheric states. Since different tracers and instruments were used in different environments, it usually has not been possible to combine data in a form most suitable for intercomparison. Furthermore, the degree of feedback between atmospheric properties that influence the disposition of pollutants and the variable pollutant loading that indirectly influences subsequent atmospheric properties has not been clearly defined.

It is hoped that tracer studies performed during the RAPS program will provide a standard basis for reducing or eliminating the costs of extensive future tracer studies for similar urban environments. For more complicated topographic regions or for the study of very localized behavior of pollutants, special tracer studies will still be required in the future, but even then the information on meaningful measurements and analyses acquired during the RAPS program for a variety of meteorological conditions will be useful. To achieve this objective, the tracer studies during RAPS will be conducted extensively as well as selectively to encompass representative meteorological conditions associated with pollution, including day-night and seasonal differences. Although periods of stormy or severe weather probably will be avoided during the tracer experiments, operations will be performed during mesoscale meteorological regimes with distinct differences in stability, wind speed, and wind direction during each season to include significant land use and energy budget variations for the urban and surrounding rural environments.

Each series of experiments will be planned to span a period of 24 hours of longer. During a 24-hour period, it is anticipated that tracer observations generally would be conducted during four periods of up to one hour each, but allowance should be made for a variety of release and sampling times. Over the year, the collection of data during approximately 24 different days should provide an extensive data bank. However, balloon tracers could be used routinely under all weather conditions, with or without other tracer experiments. Up to four observations per day during each midseason month would establish a good data set for comparison with other meteorological observations.

More frequent observations could be taken during interesting precipitation periods or during periods when low-level cloud motions are determined from satellite photographs.

General Requirements

The basis for a study of pollutant levels is conservation. Within any defined atmospheric volume, the totality of sources will equal the buildup or storage rate of the pollutant plus the divergence of the transport of pollutants over the volume. Consider a volume of atmosphere containing continuous internal sources of pollutants, with no upwind sources, that is sufficiently large to encompass any lateral spread of pollutants. If deposition at the lower boundary is negligible and if some upper boundary such as a thermal inversion exists, across which negligible vertical transport occurs, then the rate of buildup or loss of the total pollutant load within the volume will depend on the rate of transport by the wind across the downwind face of the volume. Thus, with the product of the estimated mixing depth and the mean flow rate throughout the atmospheric volume as a crude index, it is possible to assess the dilution potential of the atmosphere. Frequently, this sort of idealized assessment is based on information on the atmospheric flow rate at a single level.

To describe the potential rates of change of pollutant levels locally within the volume and to arrive at a more reliable estimate of total downwind transport, it is also necessary to describe the mixing capability (horizontally and vertically) of the atmosphere. The diffusive ability can be described in terms of the intensity of turbulence that is usually expressed by the normalized variance of the fluctuations of components of the wind velocity. Alternatively, the fluctuations of horizontal and vertical wind directions can be used as indices of turbulence. Because of the local and sometimes inconvenient nature of turbulence measurements, it is customary to describe the mixing capability of the atmosphere in terms of the stability, say, as a gradient Richardson number, which includes measures of the vertical gradients of temperature and horizontal wind speed. A somewhat better option is to obtain measures of both the stability and the related intensity of turbulence.

In summary, the essential meteorological ingredients in a pollution assessment program are simply (1) the depth of the mixing layer, (2) the distribution (especially vertical) of the wind velocity within the mixing layer, and (3) the mixing capability within the atmospheric volume. All three factors are related to the thermodynamic stability and the surface roughness, both geometric and aerodynamic. Inasmuch as steady state conditions usually are not realized in the boundary layer, it is essential also to obtain information on (4) the rates of change of stability, flow rate, and associated turbulence. The latter quantities, when not governed by larger scale synoptic changes, are related to local advection and the spatial distribution of the time-varying surface heat budget. Characteristic synoptic parameters include the gradient wind velocity at the top of the mixing layer, the large-scale stability and vertical motion, and cloudiness. The importance of all these factors must be considered in a complete analysis of data collected during the tracer and transport experiments; deposition and scavenging must be considered also where they apply to specific experiments.

Prime interest is in the ground level concentrations of pollutants, arising both from surface and elevated (tall stack) sources, at locations within the urban area and at rural locations at long distances (up to 150 km) downrange from urban sources. For the dispersion of pollutants from elevated sources and the long range transports, it is important to consider vertical dispersion rates and their changes. Furthermore, it is the vertical dispersion rate that the urban environment influences the most, but only limited information on vertical dispersion rates is available from previous studies. In fact, conclusive evidence of significant urban influences on atmospheric dispersion rates has not been documented except for enhanced dispersion at short ranges (less than about one-half mile). On the other hand, changes in meteorological conditions over the urban area have been observed consistently. Most notable are the changes in the temperature distribution and the stability, especially at night. These changes can be attributed largely to the increased storage of energy during the daytime (in addition to manmade heat sources) within the urban subsurface, with subsequent increases in thermal emission and vertical exchange at night.

The most commonly used dispersion model is the Gaussian plume formulation,

$$\frac{\bar{\chi}}{Q} = \frac{1}{2\pi\sigma_y\sigma_z\bar{u}} \exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left\{\exp\left[-\frac{(z-h)^2}{2\sigma_z^2}\right] + \exp\left[-\frac{(z+h)^2}{2\sigma_z^2}\right]\right\} . \tag{III-20a}$$

When sampling receptors are located at height $Z = 0$ this expression reduces to

$$\frac{\bar{\chi}}{Q} = \frac{1}{\pi\sigma_y\sigma_z\bar{u}} \exp\left[-\frac{y^2}{2\sigma_y^2} - \frac{h^2}{2\sigma_z^2}\right] , \tag{III-20b}$$

where Q is the continuous source strength, $\bar{\chi}$ is the average concentration, $\bar{u}$ is the mean plume transport wind, y is the lateral departure from the time mean axis (x) of the traveling plume, h is the elevation of the source above the receptor plane, σ_y is the standard deviation of the horizontal (lateral) concentration distribution within the continuous plume, and σ_z is the standard deviation of the vertical concentration distribution within the plume. Although this model implicitly assumes for a given atmospheric stability and surface aerodynamic roughness that σ_y and σ_z are only functions of x, it would be instructive, under different conditions, to examine the possible variability of σ_y with z and the variability of σ_z with both h and y.

Important factors to consider when checking models of computed concentrations are the variations in σ_y and σ_z and the relationship between them. Accordingly, these factors have been given emphasis in the recommended design considerations for tracer studies during RAPS.

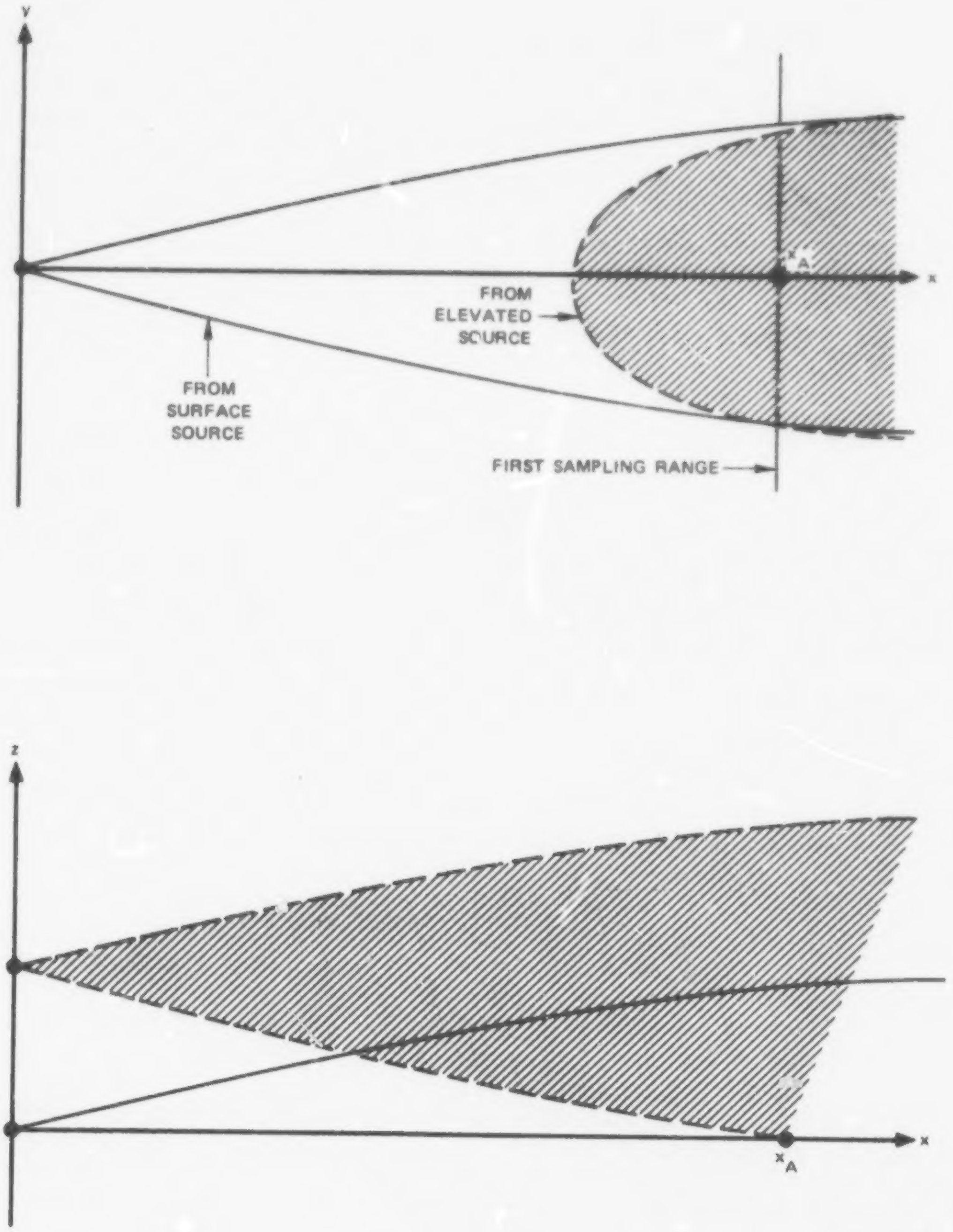
Experimental Design Considerations

Gaseous and Particle Tracers (Short Range)--With occurrence of meteorological conditions considered to be favorable for a tracer experiment, the deployment and preparation of tracer sources and sampling array are conducted on the basis of an initial low-level tetroon trajectory obtained about six hours in advance. Continuous point sources will be positioned near the surface and at one or more levels up to a height corresponding to a tall stack. A suitable platform for the elevated source(s) might be a stack itself, an available tower, or a mast mounted on the roof of a building (if no serious problem exists with local circulation anomalies). When surface and elevated sources are operated together, they will be located at the same position.

Downwind sampling of concentrations of tracer material arriving at the surface. either from surface or elevated sources, will be conducted over sampler arrays distributed in equal azimuth increments along crosswind arcs at several ranges. (A maximum range for fluorescent particle tracers will probably be restricted to distances of less than 15 km.) It is highly desirable that two or more distinguishable tracers (e.g., particles with different fluorescent colors) be released simultaneously from two or more elevations or locations and also sampled simultaneously at the same receptor location.

The most general set of tracer runs will yield data on the variation of σ_y with range x and height z (see Eq. III-1). For this experiment, various tracers will be released simultaneously from a surface and an elevated source(s); surface receptors downwind will sample the tracers simultaneously along crosswind arcs at different ranges (as shown in Figure III-8). Measures of the standard deviation of the crosswind distribution (from the various sources) at the several arc locations describe the variation of σ_y with x and h for the existing meteorological and surface conditions along the plume trajectory over the urban area. Differences in the actual dosages from the two sources will depend on the vertical dispersion rates.

With measurements of the appropriate mean transport winds, it is possible to use measurements of σ_y to determine the σ_z's for each source from Equation III-1. Results could be assessed with the aid of observed vertical and temporal variations in stability. Differences in the measured σ_y's for each source would be attributable primarily to vertical variations in σ_y. Since σ_y is normally considered to be independent of height, these measurements should be extremely valuable, especially when samples are compared at several arcs sufficiently distant so that both



SA-1365-25

FIGURE III-8 HYPOTHETICAL LATERAL-LONGITUDINAL AND VERTICAL-LONGITUDINAL CROSS SECTIONS SHOWING ENVELOPES FOR PLUMES GENERATED SIMULTANEOUSLY BY IDENTICAL POINT SOURCES AT TWO ELEVATIONS. Plume axes parallel to x axes.

plumes are within regimes for which range variations of σ_z are likely to be equal. Observed differences in the horizontal variations in the σ_z's inferred for each plume would be documented for further evaluation.

The standard deviation of the vertical distribution of material in a plume is of special interest, because it is most likely to be influenced by the urban environment. It is anticipated that pronounced changes in vertical dispersion parameters are very responsive to changes in stability and roughness. However, it is a difficult quantity to measure; in many previous experiments, it was estimated from other data rather than measured directly. The basic problem with measurements of σ_z (or with direct measurements of vertical variations in σ_y) is the difficulty of sampling in a vertical array from the surface to some distances above tall stacks and in the appropriate downwind position from a continuous source. In this critical region of the urban environment, aircraft cannot be used for sampling even if they did not influence the dispersion measurements themselves. At higher elevations in the boundary layer, meaningful dispersion data can be acquired more simply from series of tetroon flights (as described below).

Another problem with measurements of vertical dispersion near the ground is the necessity of correction for boundary "reflections." In the simplified model, a hypothetical mirror-image source is assumed to be operating below a uniformly smooth ground surface that is treated as being completely transparent. Thus, when the actual plume intercepts the surface, downwind concentrations above the lower boundary are enhanced by an amount equivalent to the additional effluent from the hypothetical source. This model (see Eq. III-1) does not allow for local irregularities in surface roughness or flow characteristics; furthermore, scavenging and settling processes are not considered in Equation III-1.

A local range study of the vertical dispersion could be conducted at and near one of the basic 30-meter observation towers. Wind, temperature, and component wind fluctuation measurements on the tower would be provided at the three standard heights and at two or more additional levels. Also, the tower could be rigged for tracer sampling along the vertical. A semimobile tower of the same height and type would be situated downwind with a dense vertical array of receptors. If the plume sources from two heights are located sufficiently close so that the towers extend through the plume tops, measurements of $\chi(z)$ and σ_z can be obtained conveniently at the two ranges. This arrangement would permit evaluation of the mean transport wind concept. Measures of σ_y could be obtained from the crosswind sampling arcs near the surface, and σ_z's could be computed for comparison with the measured σ_z's, as a function of source height h. Finally, the σ_z's could be compared with the observed Richardson numbers, whereas both the σ_z's and the Richardson numbers could be compared with the measurements of the component wind velocity fluctuations.

Direct sampling of the vertical distribution of tracers on the larger scale can be accomplished (with difficulty) from an array of samplers at different heights along the cable of a barrage balloon. The barrage balloon would be located near the center of the plume, perhaps on the basis of initial sampling at the surface along a crosswind arc. Some mobility of the barrage balloon along the arc may be desirable. Alternatively, a very tall tower, such as the KMOX TV tower, could be used as the platform for the vertical sampling array. The use of the tower requires flexibility in locating the sources upwind. On the other hand, if it were possible to instrument the tower, it would be simpler to conduct the vertical sampling there than from the barrage balloon.

From the point of view of both vertical sampling position and computational models of the dispersion, it is important to ascertain whether σ_z varies with y, i.e., crosswind within the plume. Suppose that for the fixed vertical sampling array two identical sources are positioned at the same upwind range but displaced crosswind with respect to each other as shown in Figure III-9. Different tracers would be released simultaneously from the sources and sampled simultaneously along the vertical array. If the σ_z determined from one tracer differed significantly from σ_z for the other tracer, then it would appear that σ_z in Equation III-1 could not be considered to be independent of the crosswind direction. In addition, as with the small scale measurements, σ_z could be determined by simultaneous sampling along the vertical axis of simultaneous releases at two different elevations from the same range. Results would be useful for assessing the possible influence of the lower boundary on σ_z. Therefore, this experiment actually constitutes an addition to the first experiment described in conjunction with Figure III-9.

More generally, the variation of σ_z with range or with travel time is desired. Again, for a fixed single vertical array of samplers, identical sources could be located near the surface at different ranges upwind, as shown in Figure III-10. Different tracers would be released at the same rate from each source and sampled along the vertical array. The σ_z determined empirically for each tracer would provide a description of σ_z with range X. In some situations it might be possible to determine σ_z from the altitude range above the plume core only. While this tracer experiment to define σ_z as a function of range is being conducted, sampling can be carried out along the surface arcs so that

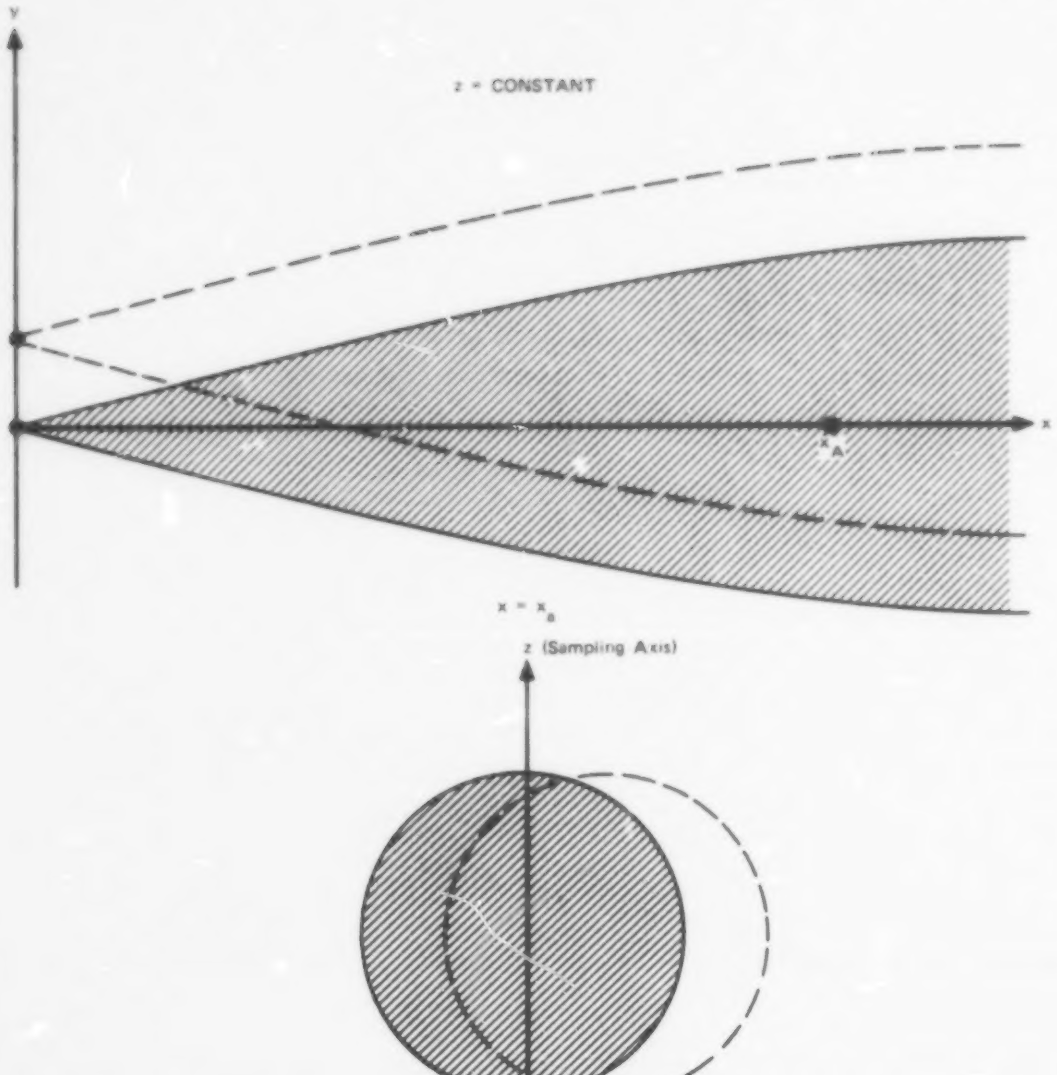


FIGURE III-9 HYPOTHETICAL LATERAL-LONGITUDINAL AND VERTICAL-LATITUDINAL CROSS SECTIONS SHOWING ENVELOPES FOR PLUMES GENERATED SIMULTANEOUSLY BY IDENTICAL POINT SOURCES DISPLACED CROSSWIND ALONG Y-AXIS

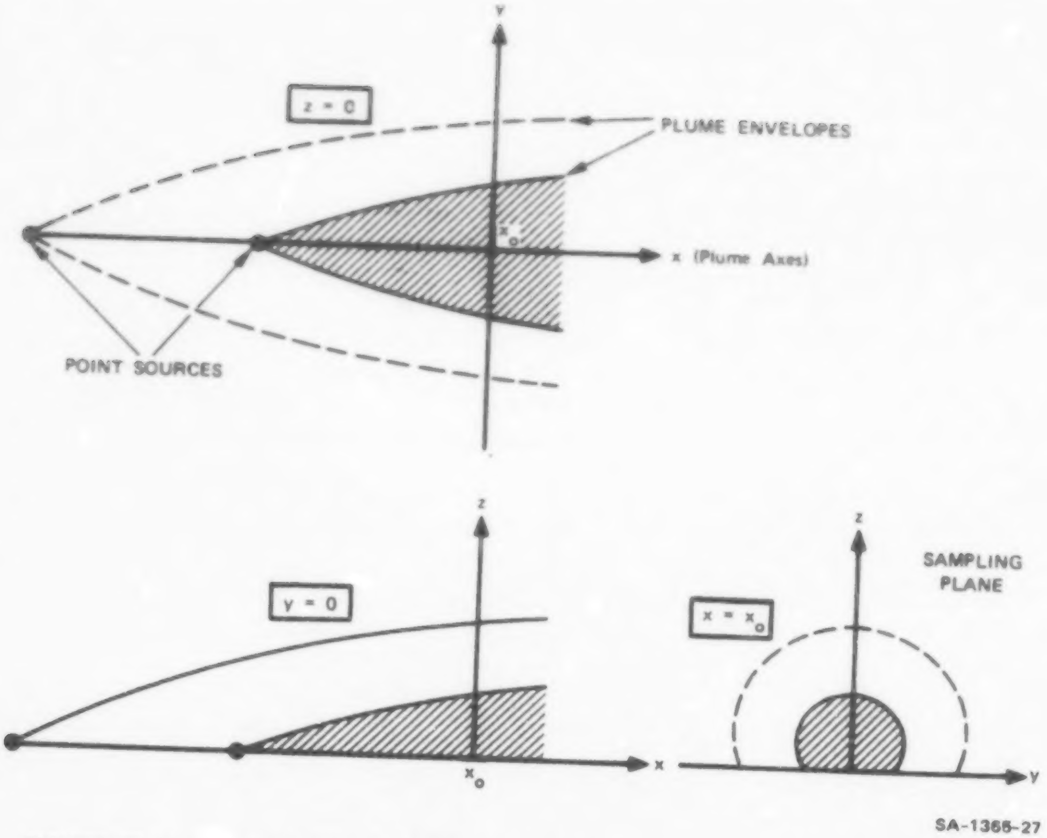


FIGURE III-10 HYPOTHETICAL SURFACE, VERTICAL-LONGITUDINAL, AND VERTICAL-LATITUDINAL CROSS SECTIONS FOR PLUMES GENERATED SIMULTANEOUSLY BY IDENTICAL POINT SOURCES AT DIFFERENT UPWIND RANGES FROM CROSSWIND AND VERTICAL SAMPLERS AT x_o

simultaneous measures of σ_y as a function of range can be obtained (as required by Eq. III-1). At the same time it might be desirable to use two vertical sampling arrays, say, the TV tower and one barrage balloon system located appropriately at a greater downwind distance. Ideally, separate surface sampling arcs would include each of the vertical sampling arrays.

With a vertical sampling array, it is also possible to determine σ_z from a series of elevated instantaneous line sources released repeatedly upwind from a helicopter.[5] However, it is not possible to make measurements of lateral dispersion with the line source. Nevertheless, the technique should be used to enable comparisons with σ_z's determined from the continuous point source experiments and with σ_z's computed from meteorological data.

Long Range Tracers--For examining the transport and dispersion of a plume along trajectories extending to about 100 km, SF_6 can be used as the tracer. This gaseous tracer will be useful for observations over the rural area downwind from the city. An elevated source might be situated in the upwind portion of the urban area in such a manner that its plume will encompass the TV tower (if it is equipped with a vertical sampling array). A vertical sampling array attached to the cable of a barrage balloon could be situated much farther downwind in the rural area, near the center of a crosswind arc of surface receptors. The vertical array probably would be necessary for very stable conditions. Measurements of σ_y and σ_z would be obtained over the rural area; these results would be compared with corresponding measurements in the urban area and with computations based on the meteorological conditions along the trajectory.

Balloon Studies--Superpressured constant-level tetroons with attached transponders can be tracked by radar or radio position finding techniques for considerable distances to provide a good representation to the three-dimensional trajectory of an air parcel (Hass, et al., 1967) for distances exceeding 100 km. The observed trajectories can provide considerable detail on the existing wind field and can be used to extend and check trajectories computed by an objective analysis of wind measurements available by conventional methods. Since tetroons actually are tracers, they also can be used to derive estimates of the diffusive capability of the atmosphere. If the release level is a few hundred meters above the ground, the resulting trajectories provide useful information, as described earlier, for positioning sampling arcs for lower level tracer studies over the urban area. In addition, each tetroon can be considered to be a platform for procuring other data. The latter concept could be easily introduced to practice by adoption of the METRAC system proposed by Control Data Corporation (1971). In this system, radio signals are transmitted from the lightweight, expendable tetroon package to an established network of receivers over the area of interest (radars are not required). The METRAC ground stations consist of a readily transportable receiving station and a simple antenna. For target positioning, a minimum of four stations must be within line-of-sight, but some redundancy in the system is strongly recommended by CDC. Within the RAPS area, it would be necessary to have receiver sites to cover adequately the longer distance transport and the several prevailing wind directions. The METRAC network could be used both for constant level wind trajectories including turbulence structure, temperature, and other compatible sensor data and for profile measurements of wind, turbulence characteristics, temperature, and other parameters using rising balloons. Accurate positioning of the tetroon as a function of time is accomplished by Doppler radio techniques, and as many as four additional measurements could be obtained and radioed separately to the receivers. Temperature and perhaps relative humidity and pressure measurements could be accommodated.

It has been demonstrated (Islitzer and Slade, 1968) that estimates of atmospheric dispersion can be obtained from individual tetroon trajectories, from successive tetroon trajectories, and from lateral distances between pairs of tetroons released nonsimultaneously. In the latter case, the time separation can be varied to obtain useful data in terms of varying atmospheric conditions. Relative diffusion estimates also can be obtained from the simultaneous release of two or more tetroons.

For the RAPS program, it would appear most profitable to operate with pairs of tetroons transmitting at different radio frequencies. The pairs could be released simultaneously at two different altitudes, and successive releasing of pairs could be performed at desired intervals. In this way it would be possible to obtain lateral and vertical dispersion estimates over the urban area and at the same time acquire measurements of stability (e.g., the gradient Richardson number) along the actual trajectory, both over the urban and rural areas. In fact, this system of observation may be the most beneficial of all tracer studies in the RAPS program.

Transformation Processes and Deposition--The source and sampling systems for the gaseous or particle tracers also will be used to conduct special observations that are designed to provide empirical data on the significance of transformation or deposition processes. For example, at select times, measurements of dispersion from specific sources using nonreactive tracer gases, i.e., SF_6, will be compared with the dispersion associated with reactive emissions, e.g., SO_2 or NO. In addition, dispersion measurements from the source using the nonreactive tracer gas (i.e., SF_6) will be compared with measurements from either unique identifiable particle emissions, e.g., heavy metals or a unique particulate tracer such as fluorescent pigment. Both types of experimental programs will be conducted during a variety of meteorological conditions.

Special Observations--It is safe to assume that most meteorological parameters that are relevant to the tracer studies will be acquired from the planned program of objective analysis of meteorological observations from surface, tower, balloon, and aircraft platforms. These will include measures of turbulence intensity in the lower 30 meters, as well as in the boundary layer above the urban structures. In addition to routine data collection techniques, remote sensing techniques will be applied to measurements of profiles of temperature, wind, turbulence, and particulate loading. Correlation spectrometer measurements of columnar variations in SO_2 and NO_2 concentrations, and lidar measurements of mixing depth are also possibilities. As described above, the recommended tetroon tracer program would itself provide valuable wind and stability data. However, additional special observations are desired to evaluate the tracer programs.

It is recommended that, at each source and at the center of each tracer sampling arc during the period of tracer release and sampling, additional measurements be made of temperature, wind speed, and lateral wind direction fluctuations. Not only will these observations complement other observations, but they also will provide a means for evaluating the applicability of simpler meteorological measurements to the techniques for estimating transport and diffusion.

The only other area requiring additional observation and/or analysis deals with the effective urban surface itself. Both the geometric and aerodynamic surface roughness (including a zero-plane wind displacement level) should be determined as a function of wind direction and range increments. This would enable evaluation of the influence of roughness variations on the wind characteristics and stability downwind. Finally, more data are required on the thermal influence of the urban surface on changes in stability. This will require representative data on the thermal forcing function (primarily solar radiation) at the surface and data on the distribution of the thermal response in terms of variations in the effective surface temperature. Such data are discussed in the following section on urban and rural radiation budget studies.

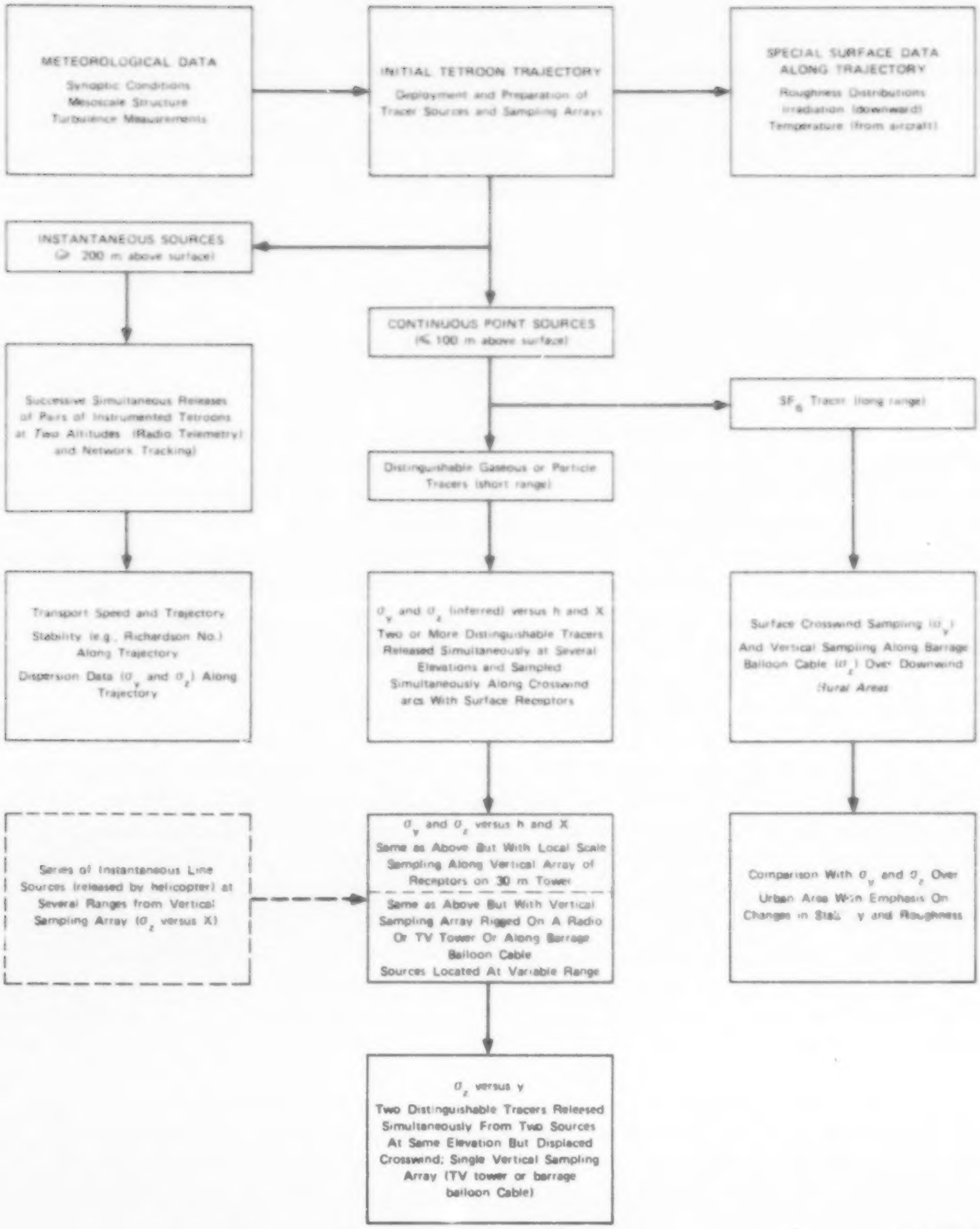
Summary

It is clear that successful tracer experiments must be correctly interfaced with the deployment of other measuring facilities, the collection and processing of other measurements, and the analysis of pertinent meteorological quantities. Ample meteorological data will be available. Figure III-11 summarizes all the basic program elements that have been recommended for the tracer experiments. Although the outline is self-explanatory, no reference is made to the extensive data analyses that are required, to the evaluation of results in terms of the meteorological parameters, or to model predictions.

The continuous point sources will be operated in the lower portion of the mixing layer, whereas the tetroons will operate generally in the middle or upper portions of the mixing layer. Of the point source plume studies near the ground, the key experiments entail the simultaneous release of individually distinguishable tracers at different heights with simultaneous sampling along the same crosswind arrays of surface receptors, and direct simultaneous measures of vertical dispersions of distinguishable plumes released simultaneously from different positions. The key tetroon tracer experiments use two tetroons released simultaneously at two altitudes with temperature sensors and radio transmission, thereby enabling the measurement of stability along the trajectory in addition to dispersion characteristics.

Urban and Rural Radiation Budget Studies

Existing observational data have revealed differences in the radiation and heat budget of the urban area with respect to the neighboring rural area. As the land use varies, differences in the radiation budgets also vary. The pollutants and differences in the boundary conditions that are responsible for the changes in the radiation budget influence the dynamics and heat budget of the lower atmosphere; in turn, feedback mechanisms exist to influence the distributions of pollutants, temperature, water vapor, and cloudiness. The effect of the urban environment


METEOROLOGICAL DATA
Synoptic Conditions
Mesoscale Structure
Turbulence Measurements
INITIAL TETROON TRAJECTORY
Deployment and Preparation of Tracer Sources and Sampling Arrays
SPECIAL SURFACE DATA ALONG TRAJECTORY
Roughness Distributions
Irradiation (downward)
Temperature (from aircraft)
INSTANTANEOUS SOURCES (≥ 200 m above surface)
CONTINUOUS POINT SOURCES (≤ 100 m above surface)
Successive Simultaneous Releases of Pairs of Instrumented Tetroons at *Two* Altitudes (Radio Telemetry) and Network Tracking)
SF$_6$ Tracer (long range)
Distinguishable Gaseous or Particle Tracers (short range)
Transport Speed and Trajectory
Stability (e.g., Richardson No.) Along Trajectory
Dispersion Data (σ_y and σ_z) Along Trajectory
σ_y and σ_z (inferred) versus h and X
Two or More Distinguishable Tracers Released Simultaneously at Several Elevations and Sampled Simultaneously Along Crosswind arcs With Surface Receptors
Surface Crosswind Sampling (σ_y)
And Vertical Sampling Along Barrage Balloon Cable (σ_z) Over Downwind *Rural Areas*
Series of Instantaneous Line Sources (released by helicopter) at Several Ranges from Vertical Sampling Array (σ_z versus X)
σ_y and σ_z versus h and X
Same as Above But With Local Scale Sampling Along Vertical Array of Receptors on 30 m Tower
Same as Above But With Vertical Sampling Array Rigged On A Radio Or TV Tower Or Along Barrage Balloon Cable
Sources Located At Variable Range
Comparison With σ_y and σ_z Over Urban Area With Emphasis On Changes in Stability and Roughness
σ_z versus y
Two Distinguishable Tracers Released Simultaneously From Two Sources At Same Elevation But Displaced Crosswind; Single Vertical Sampling Array (TV tower or barrage balloon Cable)

SA-1365-28

FIGURE III-11 PROGRAM ELEMENTS OF TRACER EXPERIMENTS

on its own climate and the climate of surrounding rural areas, as well as the significance to the larger-scale climatology, are of particular concern in future planning. Basic objectives of this portion of the experimental program are to:

(1) Document differences in urban and rural radiation budgets for the surface and boundary layer along with differences in pollutant concentrations, temperature, water vapor, and cloudiness.

(2) Provide radiation data required for the formulation of direct input into applied mathematical models of the boundary layer.

(3) Enable testing and revision of relevant radiative components currently used in mathematical models.

An observational program to document surface and near-surface differences in the radiation budgets over urban and rural areas must be conducted in conjunction with observations of temperature and water vapor profiles, careful observations of cloudiness, and measures of pollution concentrations. Because of the pronounced radiative effects of clouds, it is especially important to document differences between rural and urban cloudiness as a function of local solar time, especially if the effects of pollutants are to be deduced. As for the radiation measurements, precision instruments (Drummond, 1970) should be used because of the small relative differences to be expected.

The most important component of the surface heat budget is the available total solar radiation (a forcing function). The most significant response function is the surface temperature and the associated thermal emission from the surface. Sizable heat storage in the urban area can result from daytime solar absorption, supplemented by space heating within structures and may lead to excessive nocturnal emission. In the boundary layer above the surface, both infrared cooling and daytime solar heating influence the radiation budget; they are linked to the atmospheric stability and to the diurnally varying transport processes.

Meaningful surface measurements of the upward hemispheric flux are difficult to obtain because of the complicated surface inhomogeneities and, for the solar measurements, by complex shadowing patterns that are related to variations in sun angle. As the altitude of observation of upward flux is increased, the size of the heterogeneous surface area contributing to the measurement increases rapidly. Therefore, low altitude measurements of upward flux profiles must include spatial integration to be compatible with higher altitude measurements. On the other hand, representative measurements of downward flux can be acquired from relatively few locations with instruments situated near the upper boundary of surface structures. Since the upward flux measurements at the surface must be integrated spatially, the upward and downward flux measurements at the stations in a surface network should be obtained separately from carefully located sensors. Measurements of downward flux should be free of surface obstructions; measurements of upward flux should include representative sampling of surface materials and structures, as well as typical shadowing patterns for different sun angles.

Two quantities that are critical to a detailed analysis of the surface radiation budget are the surface albedo (as a function of sun angle) and the surface emissivity (longwave). Useful estimates of these quantities can be postulated initially on the basis of special emissivity and reflectance data for known surface materials. These estimates must be supplemented by special low-level airborne observations (and/or laboratory simulations of the surfaces with controlled sources).

The measurements of the solar irradiance at the surface should include total solar radiation (direct plus diffuse) as well as the sky (diffuse) radiation only. The latter observation will be helpful in estimating the fraction of extinction due to absorption (Lettau and Lettau, 1969). In addition, two or three spectral observations should be included to aid in subsequent analyses of observed variations. The spectral breakdown should at least include a portion of the visible spectrum that is free of molecular absorbers and a portion of the infrared spectrum that includes water vapor absorption. At the same surface sites, observations would include upward shortwave fluxes, and both upward and downward total irradiances (i.e., hemispheric fluxes, including the atmospheric or surface thermal emission). Alternatively, the longwave fluxes could be measured separately with suitably filtered radiometers.

At several key solar hour angles, aircraft observations of downward and upward solar fluxes and downward and upward total radiation (longwave fluxes included) should be made both at low level (e.g., 500 ft) and just above the boundary layer (e.g., 5000 ft). Nocturnal measurements would yield only the infrared fluxes. Although the airborne observations would not necessarily be conducted routinely over many days, sufficient flights would be conducted under differing meteorological conditions to (1) provide direct information on solar heating and infrared cooling rates for the boundary layers over urban and rural areas and (2) provide a relative measure (at the lower flight level) of the spatial distribution of the surface albedo and the thermal emission. The latter data would be used to extend the surface measurements of upward flux. During tracer experiments, the flight paths could be directed along pertinent plume trajectories. Furthermore, the aircraft data should be useful for establishing estimates of coefficients or ratios of coefficients pertaining to the aerosol scattering and absorption. (Molecular scattering and absorption components can be computed.)

It is difficult to estimate the true surface infrared emission (upward flux) from downward-looking radiometric platforms above the surface because of the absorption and emission of the intervening atmosphere. An alternative approach is to acquire airborne data on the spatial distribution of the effective surface temperature from downward-looking radiometric measurements in the 11 μm window region of the infrared spectrum and to compute a representative upward flux. Standard observations of temperature and relative humidity would enable corrections for residual absorption-emission from the atmosphere in the window region. However, since the infrared correction may depend on the concentration of particles, it might be useful to acquire infrared window measurements at the surface looking upward to note variations in atmospheric emission with variations in particle loading. Also, with the effective surface temperature for the nonhomogeneous surface area and with the observed temperature and water vapor profiles, it is possible to compute the infrared cooling distribution through the surface layer. If the radiometrically-inferred surface temperature is discontinuous with the surface temperature deduced by extrapolation of temperature profiles, the discontinuity should be maintained during the computations (unless there are obvious instrumental errors). Representative measurements of the downward infrared flux will be available near the surface, and representative measurements of both downward and upward fluxes above the boundary layer will be available occasionally from aircraft. Of particular note would be differences between observations and computations, especially when the latter are performed by neglecting the influence of particles.

The surface radiation measurements over both rural and urban areas should be conducted routinely at about the same heights above sea level, with prime consideration given to representativeness near the average height of urban structure. In general, the program outlined herein is similar to that being conducted in the Raleigh, North Carolina, area by

Flowers and Peterson. Briefly, the possible breakdown of measurements can be summarized as follows:

Surface Radiation Measurements

(1) Total direct plus diffuse solar downward

(2) Total diffuse solar downward

(3) Spectral direct plus diffuse solar downward (visible and near-infrared)

(4) Total solar upward

(5) Total thermal (long wave) downward

(6) Total thermal upward

(7) Photometer turbidity measurements

Aircraft Radiation Measurements (occasional, at two heights)

Items (1), (4), (5), and (6) above, plus measurements of effective surface temperature in the 11 μm infrared window region.

Supporting Measurements

(1) Temperature and water vapor profiles

(2) Cloudiness

(3) Pollution concentrations (near surface)

The Role of Remote Probing Techniques

Ground-based Techniques

The capability of detecting, observing, or measuring various atmospheric conditions remotely is valuable if (1) such data cannot be obtained otherwise or (2) if they can be obtained more efficiently and economically by such a capability. The basic factors are the accessibility of the portion of the atmosphere observed and the nature of the information required. Thus, although it is readily possible to measure atmospheric temperature at a height of 1000 feet given a tower, regular radiosonde, tethered balloon, or aircraft facilities, much more convenient and economical techniques of making such a measurement remotely from the surface are desirable. On the other hand, the measurement of some parameters is feasible only with remote, or at least extended path, techniques. Atmospheric transmission is a case in point, and mean concentrations of, say, CO, over extended volumes or the mean air flow across an area, are other examples.

In the present context, then, we consider what techniques of remote probing should be considered in terms of their ability to provide information more effectively and economically on (1) meteorological factors and (2) pollutants than is possible with point or in-situ measurements.

The information required is that serving both the needs of the Regional Study's objectives, such as research and model verification, and the more general need of the real-time assessment and forecasting problems.

In addition to the distinction between meteorological and pollutant measurements, it is convenient to consider the possibilities under two headings:

(1) Data collection at a Regional Study facility

(2) Research activities as part of the Regional Study.

Under (1), it is apparent that remote techniques could make very valuable contributions to the data collection networks of the Study facility. Particularly since much of the data required in studying the transport and diffusion of pollutants on the regional scale are best considered in broader and more comprehensive terms than are easily acquired by point measurements, many forms of remote measurements (e.g., long path) would provide data on the appropriate form. Again, since the three-dimensional aspects are so important it would be valuable to have more comprehensive data (in time and space) of the stability conditions and wind structure of the lower 3 km or so, than can be provided even by radiosonde and aircraft technique (or even the towers at the lowest levels). Similarly, it would be most useful to be able to determine directly the broad distribution of gaseous pollutants and their flux within an extended area rather than to have to infer such information from point measurements. (Tables III-5a and 5b show the range of such techniques.)

Table III-5a

ATMOSPHERIC CONDITIONS

Type of Observation	Purpose	Candidate Techniques
Vertical temperature profile	To assess and predict mixing depth - i.e., the depth of the neutral or unstable layer in which pollutants are usually mixed. This entails: • Identifying the presence of inversions • Assessing the height of the inversion base and the thickness and intensity of such inversions • Measuring lapse rates in the boundary layer	Microwave or IR radiometry Acoustic sounding Raman lidar
Vertical humidity profile	To aid in forecasting changes in stability and mixing depth (see vertical temperature profile)	Differential lidar absorption spectroscopy Raman lidar Microwave or IR radiometry
Vertical turbidity profile (here considered as an indication of mixing depth and stability)	To assess and predict mixing depth and monitor mixing processing	Lidar
Horizontal turbidity profiles or path transmittances	To assess "visibility" as a meteorological parameter and pollution state	Lidar and laser transmissiometry
Wind velocity, vertical profile	Input to transport and diffusion models	Acoustic sounding Lidar correlation anemometry--Doppler radar, and Doppler lidar
Vertical turbulence profile	Input and transport and diffusion models	Acoustic sounding Doppler lidar
Mean horizontal wind flow over extended path	Input to transport and diffusion models (as a more representative sample than point readings)	Lidar correlation anemometry
Mean path turbulence	Input to transport and diffusion models (as a more representative sample than point readings)	Optical and microwave propagation analysis

Table III-5b

POLLUTANTS

Data Required	Candidate Technique
Particulate distributions and concentrations (both horizontal and vertical)	Lidar Transmissiometry (limited spatial resolution and generally horizontal only)
Gaseous concentrations (with spatial variation in vertical or horizontal, or over extended sample path)	Long path spectroscopy (mainly IR) • Using fixed point--point paths (absorption) • Using fortuitous distant reflectors (absorption and emission) Passive spectroscopy (absorption and emission) Lidar using Raman, Resonant Raman, or fluorescence principle

Unfortunately, however, despite the considerable interest in this subject for so long, the possibility of incorporating remote sensing techniques in the basic data collection facility of the Regional Study is, with a few exceptions, probably unlikely until the latter stages of the project--i.e., after another three to four years. This follows from the fact that although a number of techniques and devices are under development at the present time, few are within range of being at the prototype trial stage. With the inevitable time lags entailed in manufacturing and marketing complex instrumentation, it thus is unlikely that fully tried, routine product devices will be available for procurement and installation as part of the basic data collection network in the main period of the study.

The few exceptions that are at a fairly advanced stage (notably, microwave radiometry [Thermasonde], lidar, acoustic sounders, and passive spectroscopic techniques) can probably be expected to make useful contributions to the data collection facility on an experimental basis and will become more routinely useful and effective as the Regional Study proceeds. Other techniques can be considered only as embryonic in terms of the main objectives of the Regional Study at this time. The investigation and trial of such techniques (and even those already in a more advanced stage) thus logically fall under the heading of research activities within the Regional Study, and programs to compare and evaluate such techniques, both between competing systems and between the conventional basic data collection facility, would be included as topics in the Research Plan.

Therefore, it is recommended that research into the use of remote sensing techniques in RAPS be considered in two stages:

(1) Experimental--in which devices that have already been procured and are under study within the current EPA monitoring research programs are made available to the RAPS program on a trial and evaluation basis. These instruments are:

  (a) SRI/EPA Mark VIII Lidar--capable of mapping particulate concentrations

  (b) At least one advanced model Thermasonde (microwave radiometer--capable of measuring thermal profiles and determining the height of the surface inversion)

  (c) COSPEC II Barringer Correlation Spectrometer--capable of determining SO_2 and NO_2 either over an extended path or in terms of total burden in the vertical

  To these should be added

  (d) A vertical acoustic sounder (of which a number of prototypes have been built)--capable of monitoring the depth of the mixing layer and identifying inversion levels. (An approach that is very attractive for operational use because of its relatively low cost.)

All these techniques have been sufficiently well developed and demonstrated to indicate that they could early provide useful inputs into RAPS observational programs--especially those concerned with special projects. The further trials and experience gained in such experimental collaborations would hopefully qualify them for more routine use in later stages of RAPS, beside providing a valuable input to the technology of remote probing in general.

(2) Research--in which devices that are less well developed at the present time or have yet to be well tested are used in

research programs to investigate their potential for more routine use. These include:

(a) Long path absorption spectrometers already procured or under procurement:

  (i) General Dynamics IR grating spectrometer (for various species over a path length of 2 miles)

  (ii) Bendix UV variable filter spectrometer (for O_3 over path lengths of 0.25 to 1.5 miles)

  (iii) General Electric CO_2 Laser (ILAMS) long path absorption spectrometer (for ammonia and ethylene and other gases over a 2-mile path)

(b) Other devices and techniques not yet procured or developed but that are likely to become available during the RAPS period:

  (i) Other types of long path or passive spectrometers

  (ii) Acoustic sounding systems for measuring temperature, humidity, and wind profiles

  (iii) Advanced microwave or IR radiometers capable of measuring temperature profiles (using angular scanning and multifrequency techniques in combination)

  (iv) Fluorescence determination of NO_2 remotely (Birnbaum et al., 1971)

  (v) Any airborne applications of such remote probing techniques

  (vi) Lidar or other optical devices for measuring wind profiles.

Satellite Applications

Major advances in monitoring atmospheric behavior on the mesoscale and synoptic scale are expected in the next five years from the use of improved weather satellites. A careful study should be made to determine whether the benefits of satellites to RAPS will warrant the expense of obtaining the data; our initial estimate is that this will be true.

Perhaps the principal application of these data would be in providing an overview of the clouds and weather systems in the area, with a resolution (in the visible spectrum) of two miles or better. From the geosynchronous satellites, such data may be obtained at approximately half-hour intervals and therefore will be up-to-date. Infrared (11 micron) sensors will give information with approximately four-mile resolution that can be used in describing surface temperature when skies are clear, or the altitude of cloud layers. Time lapse viewing of such pictures shows the cloud motions; these may be used as data for objective wind analysis.

The first SMS/GOES satellite carrying the sensors mentioned above will be launched in late 1972. Its subpoint will be over the equator at 100°W; therefore, it will have a good view of the central United States. About a year or so later, the second satellite of this series will be launched, and it will have additional sensors for measuring vertical temperature and humidity profiles. However, it should be realized that these profiles will give somewhat smoothed information in comparison to radiosondes and will not reveal the detailed structure of phenomena such as temperature inversions.

Similar sensors carried by polar orbiting satellites are also being improved. Scanning radiometers will obtain pictures in the visible and infrared with 0.5 mile resolution. Such pictures will be obtained twice a day in a given locality from the ITOS and Nimbus satellites. Several of these polar-orbiters will be launched between 1972 and 1975.

The application of these satellite data for RAPS would include:

(1) Monitoring weather systems in real time. Such systems would include highs, lows, fronts, squall lines, fog, and cyclone formation.

(2) Making mesoscale analyses of winds and temperature for use in diffusion models and air quality studies.

(3) Controlling field experiments and data gathering activities.

(4) Providing a live display for the public, illustrating the relationships between air quality and weather patterns.

These satellite data may be obtained in two different ways. One way is to obtain the data directly using a special ground receiving station. This would require an installation worth at least several hundred thousand dollars and would require a significant staff. The second alternative would be to obtain the data by facsimile or by leased lines from a direct readout site that NOAA is planning for the severe storm center at Kansas City. The associated costs would depend on the volume of data needed. Further study of this question is required.

Upper Air Sampling Program

To understand and model the dispersion of contaminants on the regional scale, routine measurements are required of the three-dimensional (space) structure of the planetary boundary layer and its temporal variations; the specific application of these data was presented in the preceding discussions on dispersion modeling. The surface based monitoring facility is designed to satisfy these requirements at the lower levels. Two primary alternatives appear to be available for in situ measurements at the upper levels: aircraft and balloon systems. The discussion on remote probing techniques presents a possible alternate approach for upper air observations. However, in the near future, the latter may be safely ruled out for routine, broad-scale usage.

The advantages and drawbacks of aircraft measurements are fairly well known. To sum a few of the more important considerations, it should be recognized that control of the measurement platform is a distinct advantage. Therefore, measurements can be made at the desired location and can be repeated in short order. On the other hand, the accuracy of the wind measurements in particular is open to question because of aerodynamic effects, and, equally important, the operating costs of the system are quite high. As such, it seems that use of an aircraft would be highly desirable for special, short-duration research studies but perhaps not for daily operation. Specific recommendations for the use of aircraft are given in Chapter XIV.

The collection of meteorological data by balloon techniques is restricted to the more or less uncontrollable trajectory of the balloon. This can be an advantage when studying Lagrangian or air parcel processes, but it is a disadvantage when objective cross sections of Eulerian conditions over the region are desired. Some flexibility is introduced in the use of both rising and constant-level balloons, especially if it is possible to simultaneously track several balloons of one or both types. In this manner, one could observe the vertical structure of the atmosphere at several locations (at the same time) while also observing the nature of wind trajectories over the region at various altitudes and/or geographical locales. Operating costs can differ widely among various balloon systems and may thus be an advantage for one and a drawback of another.

The operational flexibility and expense of the proprietary CDC METRAC system appear to be attractive features. The METRAC tracking system employs Doppler tracking techniques to determine a highly accurate three-dimensional position of an inexpensive radio transmitter. This lightweight transmitter, carried either on a rising or horizontally floating small balloon, permits fine-scale measurements of the wind and as many as four measurements from additional sensors on the balloon. After the signal is received by a minimum of four portable ground stations, its information is relayed by radio or landline communication to a central data terminal. The data are then recorded for future use or processed immediately by a small computer.

Primary METRAC advantages are low cost and high accuracy. Flight packages are expendable, as in radiosondes, and the cost is comparable or lower. Ground station costs are also much less than the usual instrumented towers, rawinsonde sounding system, or radar positioning system. The master station is fully automated, and the only operator requirement is starting the receivers before launch. Since data generated by the master station are formatted for direct computer processing, no intermediate processing preparation is required. A small computer can process an entire profile in less than five minutes. The high accuracy in three-dimensional positioning permits detailed calculation of resultant winds. This high resolution of data in time and space thus enables the determination of small-scale features of vertical or horizontal profiles.

Several other advantages are basic to the system. By using a network of these systems, the inherent accuracy and range is limited only by the boundary of the network. Thus, tracking over hundreds of kilometers is possible as long as at least four receivers remain in radio line-of-sight. Observations can be taken day or night in all weather conditions. Simultaneous tracking of multiple balloons is possible with special modification of the system. Finally, the flexibility provided by the multisensor capability should not be underestimated. Simultaneous point (single balloon) and gradient (multiple balloons) measurements of wind, temperature, humidity, pressure, and net radiation can be obtained at precisely determined locations.

The determination of space coordinates at any instant of time is based on the system's ability to monitor differential Doppler shifts between the received frequencies at the various ground-based receivers. At each ground station, a receiver translates both the NBFM telemetry information and its Doppler frequency shift from a radio frequency carrier to an audio frequency carrier signal that comes unmodulated from the central data terminal. This data-carrying signal from each receiver is then transmitted to the central data terminal via a voice channel communications link.

While a minimum of four ground stations is required, it is recommended that seven stations, arranged in a hexagonal pattern with one in the center, be used to provide some redundancy and thereby reduce the vulnerability of the system to equipment failures or multipath and other propagation effects. Use of seven stations allows the calculation of 35 coordinate positions for the mobile transmitter at any given instant. Should propagation problems affect the signal at one ground station, a total of 15 coordinate positions is available; while if the signals at two of the ground stations are affected, a total of five coordinate positions is still available.

At the data terminal, the signal from each ground station is sampled a minimum of 50 times per second. This high rate is necessary to recover the telemetry data even though the modulation deviation is low. Each balloon-borne sensor is sampled once each second; however, 50 positions per second may be computed if this fine a time scale is desired. Ordinarily, and especially in on-line computing, it is not desirable to obtain position information at this rate, and digital filtering of the data may be advantageous.

Indications are that the use of a six-balloon METRAC system could provide six 15-second averages of vector winds every 15 seconds with a resolution of better than 0.1 m/sec. Winds for a one-second average (single balloon) can be resolved to 0.3 m/sec. Therefore, the system is capable of resolving the vector velocity of the balloon to a degree equal to or better than the representativeness of the balloon in describing the ambient wind field. Table III-6 from the HMSO Handbook of Meteorological Instruments (1961) is a comparison of alternate optical and radio tracking techniques.

The ground-based receivers are housed in lightweight weatherproof packages and can be mounted on a 10-foot pole that is used to support the receiver antenna system. The pole itself is sectioned for easy portability and is supported by a guy system that also functions as a reflecting screen for the antenna. The antennas are simple and stationary, since the use of directional arrays is not required.

Table III-6

VECTOR ERROR OF WIND (m/sec) AT 5 km AS A FUNCTION OF TRACKING SYSTEM AND RATIO (Q) OF MEAN WIND TO ASCENT RATE

Q	A	B	C	D
1	0.5	0.5	0.5	1.0
2	0.5	1.0	1.0	2.0
3	0.5	2.0	1.5	3.6
4	1.0	3.6	2.5	5.1
5	1.0	5.6	3.6	7.1

A - Radar
B - Optical or radio-theodolite and radiosonde height
C - Double theodolite or direction-finder
D - Optical or radio-theodolite and assumed ascent rate

Simultaneous tracking of more than one balloon requires equipment to be added at both the receiver and data terminal sites in addition to the use of separate transmitting frequencies for the mobile transmitters. Each receiver will require an additional IF unit for each channel used as well as a multiplex unit for the data link. A multiplex adapter will also be needed at the data terminal unit in addition to either a high speed incremental tape unit or a high speed data transmission link to a computer.

Apart from the advantageous cost and high-resolution features of METRAC, the flexibility of the system is particularly attractive. For example, it would be extremely valuable to have the capability to collect five or six vertical soundings simultaneously throughout the study region at, for example, a one-hour periodicity during intensive model test periods or, say, four-to-six per day (perhaps at fewer locations) on a routine basis. The coincidental measurement of wind trajectories over the region would also provide necessary meteorological data. With a system such as METRAC, both programs could be undertaken simultaneously, or perhaps the vertical soundings could be interspersed in the horizontal tracking program.

Private communications with personnel at CDC have provided a tentative cost and time projection for implementation of an operational multiballoon, multisensor system. The basic system has been tested on a limited basis, but an additional six-month preliminary design and test period is required toward implementation as an operational system. This program would evaluate and resolve design criteria for particular space and time scales (resolution), available radio frequencies, and sensor requirements. A projected estimate for the cost of this initial program is on the order of $100,000 including hardware costs. At the end of the program, a final decision could be made on the practical feasibility of incorporating the system into RAPS. In the event the system did not satisfy its projections, a more conventional system could be brought in with little or no tool-up time required.

If METRAC does prove itself as a result of the test period, an additional outlay of about $250,000 would be necessary over the following year to finalize software and station designs, fabricate the system, locate and prepare the sites, and install the receivers. This cost is for a multiple-balloon system; a single balloon system might save only 10%. Hardware costs would be additional: central station, $50,000; slave (remote) stations $1500 to $2000 each; small computer, $30,000 to $50,000. For the St. Louis region, approximately 18 slave stations would be required to adequately track up to six packages simultaneously to a minimum range of 150 km. Station hardware costs for a single-balloon system would be approximately half of that.

In summary, the approximate total costs for the 18-month efforts for single versus multiple-balloon systems are: (1) single-balloon tracking capability for one central and six remote stations, including computer and all design costs, is $395,000; and (2) six-balloon tracking capability for one central and 18 remote stations, including computer and all design costs, is $475,000.

As an example of the operating costs of an alternate system, the NOAA radar-tracking tetroon system is estimated to cost in the neighborhood of $100,000 for one month of intensive operation and subsequent data processing. In fact, these data are not routinely available in real time and require several to be fully processed. METRAC could be expected to substantially reduce this operating cost to something on the order of

$8000 (or less) per release location for the weighted manpower costs for an intensive (round-the-clock) operation for one month; data processing in real-time is included in the operation and costing schedule.

In summary, it is strongly recommended that the feasibility of a system such as METRAC be explored for the routine collection of upper level wind, temperature, humidity, and pressure data. Funding for upper air measurements in the RAPS program has been based on the above listed METRAC costs for the multiple-balloon tracking system.

Plume Dispersion Studies

The various aspects of the dispersion of a plume from a point source should be studied within the framework of the regional program for a number of reasons. The most obvious reason is that the study of plume dispersion is an integral part of the small-scale or localized air quality modeling and control program. Indeed, it is the large point source that can have an overwhelming influence on the level of air quality of localized areas even though its effect on regional air quality may be minimal. Careful treatment of the strength and location of the source, as well as of the dispersion of the plume downwind, is required in the development of regional emissions grids. Another important area for study is the nature of long range plume dispersion, particularly from elevated sources under stable atmospheric conditions or when a capping inversion is present aloft. In these cases, the air quality of a given region may be influenced severely by emissions from adjacent regions. As an example, the characteristic reddish pall of Gary, Indiana, can sometimes be seen at Madison, Wisconsin (about 250 km distant), under "favorable" atmospheric conditions. Another ramification is the varied hue of the sunset on approaching the Northeast megalopolis from the sea.

Therefore, plume studies constitute a part of the Regional Study in a number of ways: (1) through the evaluation of the impact of local sources on both local and regional air quality observations and modeling, (2) as basic or special studies directly concerned with the program of detailed model evaluation, and (3) in determining the ramifications of long range plume dispersion on regional air quality. Item (3) must not of necessity be restricted to the St. Louis study region but would be worthwhile in view of the wealth of additional data and special facilities to be available there.

Plume-related studies may conveniently be classified into three physically distinct areas, namely the physical processes related to the initial behavior of the plume after it exits the stack, the transformation and removal processes affecting the pollutants, as well as their transport and diffusion. Studies of the initial rise and dispersion of the plume should seek to further verify the various available plume rise formulations (Briggs, 1969) and to parameterize where necessary the meteorological and engineering input requirements.

Three processes affecting initial plume behavior should be considered: (1) rise of the plume relative to the ambient vector wind due to the initial buoyancy and inertia of the efflux, as well as the effects of the release of latent heat through plume condensation; (2) aerodynamic effects due to nearby buildings and topographical features, in addition to effects of the stack itself; and (3) diffusion by turbulence of the ambient air. Intensive investigations on the nature of aerodynamic effects in general need not be of concern to RAPS because of their extremely localized behavior as in the case of structural effects; furthermore, these can be eliminated with proper design before construction. Topographical (aerodynamic) effects may be of wider applicability, although the geography of the St. Louis region may not permit or warrant research in this area. Briggs (1969) suggests that additional effort be expended in defining the role that atmospheric turbulence plays in the initial rise of the plume through its effect on entrainment. Recent attention has been given to the role that water vapor condensation plays in affecting plume buoyancy (e.g., Csanady, 1971, and Wigley, 1971); the effect may be significant in some cases, since the latent heat released through condensation may on occasion equal the sensible heat of the efflux. Additional research in the way of special studies seems desirable in this area. Another area where research may be warranted is defining the significance of radiative effects (both absorption of shortwave radiation and emission of longwave radiation) on plume behavior. Transformation and removal process can affect both the initial plume rise and subsequent concentrations downwind; their study within RAPS is discussed in Chapters IV and V.

Short and intermediate range studies of the dispersion of plumes from elevated point sources fall partly within the scope of the tracer programs discussed earlier in this chapter. An alternate program for the study of plume behavior over the short to long range scales is through the use of mobile, remote probing techniques. Two systems appear applicable, and they may be considered for both joint and independent usage; they are the correlation spectrometer and the lidar. The former can provide data on the mass loading of a particular gas in a vertical column, while the latter can give information on the relative mass distribution of particles in one dimension with a single "shot" and in two or three dimensions by scanning and/or sweeping.

An experimental program should consider the use of more than one instrument of each type. One configuration would use a lidar and a correlation spectrometer at each of two locations along the plume trajectory. Cross-plume sections at the two ranges by the spectrometer would provide data for the study of transformation and removal processes as a function of time or distance. The data might also be useful for computing absolute concentrations from the "relative" lidar returns. The lidar, in turn, can provide information on the detailed structure of the plume cross section, thereby permitting the evaluation of lateral and vertical diffusive processes and also their dependence on height and lateral distance, from the time-averaged returns. Maximum advantage can also be made from the data available from the routine meteorological and air quality facility of the RAPS program. For example, a set of portable METRAC slave stations should be used at the site to collect frequent vertical wind, temperature, and humidity profiles at one or more points along the plume trajectory. Additionally, a series of constant-level balloons should be employed to document the mean wind trajectory and diffusion characteristics at one or more heights (as dictated by the height of the plume centerline). Furthermore, a helicopter-borne lidar system could be used to study Lagrangian diffusion of the plume by following the tetroons. Other possibilities for experimental design would incorporate, say, an airborne lidar together with a surface-based system, and so forth; such refinements would be a logical extension of the multiprobe experiment outlined here.

Studies of Spatial Variabilities

In the analysis of the sensitivity of models, the objective is to evaluate the response of the model output to variations in its input parameters. For convenience, these variations have often been termed errors and sometimes qualified as either random or systematic. In practice, these "errors" may arise from a variety of sources: (1) incorrect measurement or instrument error, (2) unrepresentative siting of the sensors (such as too close to an obstruction for a wind measurement), and (3) natural variability of the parameter over the region of interest. Similar variations or errors can exist between an observed measure of air quality and a value truly representative of conditions over a desired area or volume. Therefore, a model with a 1 km^2 spatial resolution requires input data representative of conditions on this scale and, similarly, representative air quality observations and/or the degree of ambient variability with which its computations may effectively be verified. Some models, such as the SRI model described earlier, have attempted to describe subgrid variations, but have only been successful because special representative measurements were made; in this case the time-averaged subgrid spatial variations often exceeded the mean grid value.

Therefore, it is recommended that a research task be delineated with the objective of defining the spatial (on various scales according to the simulation resolution that is required) variability of meteorological and air quality conditions. Furthermore, the program should seek to explicitly parameterize these variations on the basis of readily available physical characteristics, such as the strength and distribution of emission sources, atmospheric stability, wind speed, and the surface aerodynamic roughness and zero-plane displacement height. Another objective of the program should be to investigate simultaneously the analogous resolution required for temporal considerations and the joint time-space criteria.

The hardware facility of the RAPS program has been designed to investigate these problems, among others, in addition to providing baseline and routine model verification data. The basic grid of Class A meteorological and air quality (tower) stations, together with the objective analysis techniques discussed earlier, will provide data for the evaluation of the significance of subregional scale features under various weather types. Questions such as the parameter resolution that is required as a function of distance from a receptor can be answered, as well as the applicability of the assumption of steady-state conditions over particular averaging times. The upper air (balloon and aircraft) programs will complement the surface facility for evaluation of the significance of four-dimensional boundary layer structure toward making predictions of ground level air quality or conditions, say, in the "building height" zone.

Primary emphasis of the RAPS program, however, is on the simulation and ultimate control of near-surface air quality. It is in this light that the flexibility of the Class C, transportable station network will be of extreme value in general and for the study of local variability in particular. If it is assumed that, as an example, a one-kilometer square represents the areal scale that is required for a particular application, then a series of experiments should be designed as outlined below. To summarize, the degree of variability of air quality over the 1-km square is controlled by three independent features: (1) the state of the atmosphere, (2) the nature of the emissions field, and (3) geographical characteristics. In a controlled laboratory experiment, the approach to solution of the problem would be to fix two of the three features at any given time and to study the effects of variations of the third, subsequently altering and repeating the scheme until the effects of all combinations of features had been evaluated. Unfortunately, the real world does not cooperate in providing these distinct combinations. For a given region, geographical and emissions features are usually, and for all practical purposes, fixed (not implying stationarity of emissions sources). The third feature, the state of the atmosphere, does vary although not always in a systematic or predictable fashion. Still, distinct meteorological regimes (as described, for example, by the Pasquill-Gifford stratifications) do exist and can be encountered throughout the year in St. Louis. The general climatology for the region is discussed in Chapter XV, while some specific data on the frequency of air pollution-potential conditions are given in Chapter XI. Therefore, the emissions and geographical features of the St. Louis region must be examined on a 1-km grid mesh, and a number of squares selected that are representative of reasonably distinct combinations of varied features. First, the geographical features should be examined and stratified by their average values of aerodynamic roughness (z_0) and, possibly, displacement height (d). For example, combinations of z_0 values for the ranges 0-10 cm, 10 cm-1 m, and > 1 m, and d values of 0-1 m, 1-5 m, and > 5 m might be chosen and would result in nine geographical categories. Distribution of the emissions may not be as straightforward and might follow in the first approximation simply by considerations of emission type (i.e., the type of pollutant) and strength (say, moderate or strong). Thus for a given pollutant, a possible total of 18 emission-geographic features may need to be studied under the six stability categories. To evaluate all combinations would be an endless and unnecessary task; priorities will be assigned by the frequency and physical importance of the many types and only selected combinations studied.

Having chosen the areas for study, the network of some 20 transportable stations would be systematically deployed over the region to provide an objective analysis of spatial (subgrid) and temporal variations of meteorological and air quality conditions. Each area would thus provide a statistical indication of the variabilities through stratification by the atmospheric conditions. The length of time that a study might require in each area will depend on the frequency of occurrence of the weather types. The length of time will also depend on the time of the year and to a lesser degree the local environment (i.e., rural or urban). Data of this type can be obtained from the Environmental Data Service at the National Climatic Center under the "Star Program," where the seasonal and annual distribution of stability category (by wind direction) is tabulated for five-year periods at various cities. These data should be obtained for St. Louis (if available) and used to construct a reasonable operational timetable for these (and other) experiments. However, a one- to two-month minimum should be anticipated for each area.

REFERENCES

American Meteorological Society, Conference on Air Pollution Meteorology, April 5-9, 1971, Raleigh, North Carolina

Belmont, A. D., Personal communication, Control Data Corporation, Research Division, Minneapolis, Minnesota (1971)

Birnbaum M., et al., "Fluorescence Determination of Atmospheric Pollutants, NO_2," paper presented at Joint Conference on Sensing Pollutants, Palo Alto, California, November 1971

Briggs, G. A., Plume Rise, U.S. Atomic Energy Commission, Division of Technical Information, 81 pp. and vi, 1969

Control Data Corporation, Minneapolis, Minnesota, private communication (1971)

Corrsin, S., Outline of some topics in homogeneous turbulent flow, J. Geophys. Res. 64(12), 1959, pp 2134-2150

Cramer, H. E., A practical method for estimating the dispersal of atmospheric contaminants, Proceedings of 1st Nat'l. Conf. on Applied Meteorology, pp. C33-C55, American Meteorological Society, Hartford, Connecticut, 1957

Cressman, G. P., An operational objective analysis system, Monthly Weather Review, 87 (10), 1959, pp. 367-374

Csanady, G. T., Bent-over vapour plumes, presented at conference on Air Pollution Meteorology, April 5-9, 1971, Raleigh, North Carolina

Dabberdt, W. F., Wind and Turbulence Structure in the Boundary Layer over the Antarctic Plateau, Ph.D. Dissertation, Met. Dept., U. Wisconsin, Madison (1969)

Drummond, A. J. (ed), Precision Radiometry, Advances in Geophysics, Vol. 14, Academic Press, New York and London, 1970

Edinger, J. G., Changes in the depth of the marine layer over the Los Angeles Basin, J. of Meteorology, 16(3), 1959, pp. 219-226

Edinger, J. G., and R. Helvey, The San Fernando convergence zone, *Bull. Amer. Meteorological Society*, *42*(9), 1961, pp. 626-627

Endlich, R. M., and R. L. Mancuso, 1968: Objective analysis of environmental conditions associated with severe thunderstorms and tornadoes, *Monthly Weather Review*, *96*(6), pp. 342-350

Eschenroeder, A. Q. and J. R. Martinez, 1969: Mathematical Modeling of Photochemical Smog, General Research Corp., Santa Barbara, California, IMR 1210

Flowers, E., and J. Peterson, personal communication, Environmental Protection Agency, Research Triangle Park, North Carolina, 1971

Gifford, F. A., An outline of theories of diffusion in the lower layers of the atmosphere, *Meteorology and Atomic Energy: 1968*, U.S. Atomic Energy Commission, Division of Technical Information, 1968

__________, Use of routine observations for estimating atmospheric dispersion, *Nuclear Safety*, *2*(4), 1961, pp. 47-51

Gifford, F. A., Jr., Atmospheric diffusion in an urban area. Paper presented at the 2nd IRPA Conference, Brighton, England, 1970

Gifford, F. A., Jr. and Steven R. Hanna, Urban air pollution modeling. Paper presented at International Air Pollution Conference of the International Union of Air Pollution Prevention Associations, 1970

Gerrity, J. P., A physical-numerical model for the prediction of synoptic-scale low cloudiness, *Monthly Weather Review*, *95*(5), 1967, pp. 261-282

Hadeen, Kenneth D., AFGWC boundary layer model. AFGWC Technical Memorandum 70-5. AFGWC Air Weather Service (MAC), Offutt AFB, Nebraska, 1970

Handbook of Meteorological Instruments, Instruments for upper air measurements. London: Her Majesty's Stationery Office, M. O. 577, Air Ministry, Meteorological Office, 1961

Hanna, Steven R.: Simple methods of calculating dispersion from urban area sources. Presented at the Conference on Air Pollution Meteorology, Raleigh, North Carolina, sponsored by the American Meteorological Society (April 1971)

Hass, W. A., et al., Analysis of Low-Level Constant Volume Balloon (Tetroon) Flights over New York City, *Quart. J. Roy Meteor. Soc.*, *93*, 398, 483-493 (1967)

Hay, J. S., and F. Pasquill, 1959: Diffusion from a continuous source in relation to the spectrum and scale of turbulence, *Advances in Geophysics*, *Vol. 6*, Academic Press, New York, 1959, pp. 345-365

Hilst, Glenn R., Sensitivities of Air Quality Prediction to Input Errors and Uncertainties, Proceedings on Multiple-Source Urban Diffusion Models, Editor: Arthur C. Stern, U.S. Environmental Protection Agency, Air Pollution Control, Research Triangle Park, North Carolina, 1970, pp. 8-1 to 8-40

Hilst, G. R., and N. E. Bowne, A Study of the Diffusion of Aerosols Released from Aerial Line Sources Upwind of an Urban Complex, Volumes I and II, Final Report, IV 025001A 128, Travelers Research Center, Inc. Hartford, Connecticut (1966)

Hilst, G., J. Yocum, and N. Bowne, The development of a simulation model for air pollution over Connecticut, Vol. 1, Summary Report 7233-279a, The Travelers Research Center, Inc., Hartford, Connecticut, 1967

Islitzer, N. F., and D. H. Slade, Diffusion and Transport Experiments, Chapter 4, *Meteorology and Atomic Energy 1968*, U.S. Atomic Energy Commission, Division of Technical Information, 1968

Johnson, W. B., et al., Field study for initial evaluation of an urban diffusion model for carbon monoxide, Comprehensive Report, Coordinating Research Council CAPA-3-68(1-69), Environmental Protection Agency, Stanford Research Institute, Menlo Park, California, 215 pp and XXV, NTIS No. PB-203-469, 1971

Kerlin, James, AFGWC mesoscale prediction model. AFGWC Technical Memorandum 70-1, Air Force Global Weather Central, Air Weather Service (Mac), Offutt AFB, Nebraska, 1970

Lamb, R., An Air Pollution Model of Los Angeles, Master's thesis, UCLA, 1968, 104 pp. and viii.

Leighton, P. A., and R. B. Dittmar, Behavior of aerosol clouds within cities, Joint Quarterly Report No. 2, October-December 1952, Contracts DA-18-064-cmc-1856 and DA-18-064-cml-2282, Stanford University and Ralph M. Parsons Co., 100 pp., DDC No. AD 7261, 1952

__________, ibid., Joint Quarterly Report No. 4, January-March 1953, 218 pp., DDC No. AD 31509, 1953

__________, ibid., Joint Quarterly Report No. 4, April-June 1953, 196 pp., DDC No. AD 31508, 1953

__________, ibid., Joint Quarterly Report No. 5, July-September, 238 pp., DDC No. AD 31507, 1953

__________, ibid., Joint Quarterly Report No. 6, Vol. I, October-December 1953, 246 pp., DDC No. AD 31510, 1953

__________, ibid., Joint Quarterly Report No. 6, Vol. II, October-December 1953, 187 pp., DDC No. AD 31511, 1953

Lettau, H. H., A new hypothesis for the relationship between eddy and mean states, Physics of Fluids (Supplement 1967), S79-S83, 1967

__________, A new vorticity-transfer hypothesis of turbulence theory, J. Atm. Sc., 21, 453-456, 1964

__________, Longitudinal Versus Lateral Eddy Length-Scale, J. Atm. Sc., 23, 151-158, 1966

__________, Physical and meteorological basis for mathematical models of urban diffusion processes, presented at Symposium on Multiple-Source Urban Diffusion Models, Chapel Hill, North Carolina (October 1970)

__________, Shortwave Radiation Climatonomy, Tellus XXI, 1, pp. 1-14, 1969

__________, Synthetic Climatology, Berichte des Deutschen Wetterdienstes, No. 38, Bad Kissingen, Germany, 1952

__________, Three-dimensional Turbulence in Unidirectional Mean Flow, Annual Reports 1966-1967, Contract No. DA-AMC-28-043-66-G24, Dept. of Met., Univ. of Wis., 127-156, 1968

Ludwig, F. L., Urban air temperatures and their relation to extraurban meteorological measurements, January 1970 Meeting of Amer. Soc. Heat., Refrig., and Vent. Eng., San Francisco, 1970

__________, Urban Temperature Fields, presented at WMO Symposium on Urban Climates and Building Climatology, Brussels, 15-25, October 1968

Ludwig, F. L., et al., A practical multipurpose urban diffusion model for carbon monoxide, Final Report, Coordinating Research Council Contract CAPA-3-68, National Air Pollution Control Administration Contract CPA 22-69-64, Stanford Research Institute, Menlo Park, California, 184 pp., National Technical Information Service No. PB 196003, 1970

MacCracken, M.C., et al., Development of a multibox air pollution model and initial verification for the San Francisco Bay area, presented to 52nd Annual Meeting of American Meteorological Society, January 10-13, New Orleans, Louisiana, 1970

Manabe, S., et al., Tropical circulation in a time-integration of a global model of the atmosphere, *J. Atm. Sc.*, *27*(4), pp. 580-613

McElroy, James L. and Francis Pooler, Jr., St. Louis dispersion study Volume II-analysis. Consumer Protection and Environmental Health Service, National Air Pollution Control Administration, Arlington, Virginia (1968)

__________, St. Louis Dispersion Study, Volumes I and II, U.S. Department of Health, Education, and Welfare, National Air Pollution Control Administration, Arlington, Virginia, 1968

Meade, P. S., The effects of meteorological factors on the dispersion of airborne material, *Rassegna Internazionale Elettronica e Nucleare*, 6 Rassegna, Rome, July 1959, Atti del Congresso Scientifico, Sezione, Nucleare, 1959

Michael, A., The effects of wind and turbulence profiles upon atmospheric dispersions, presented to Conference on Air Pollution Meteorology, April 5-9, Raleigh, North Carolina, 1971

Moses, Harry, 1969: Mathematical urban air pollution models. ANL/ES-RPY-001. Argonne National Laboratory, Argonne, Illinois, 1969

Neiburger, M., The role of meteorology in the study and control of air pollution, Bull. Amer. Meteorol. Soc., Vol. 50, No. 12, 1969, pp. 957-965

Pack, D. H., and J. K. Angell, A Preliminary Study of Air Trajectories in the Los Angeles Basin as Derived from Tetroon Flights, Monthly Weather Review, 91: 583-604, 1963

Pandolfo, J. P., et al., Prediction by numerical models of transport and diffusion in an urban boundary layer, Final Report, Contract CPA 70-62, Environmental Protection Agency, by The Center for the Environment and Man, Inc., Hartford, Connecticut, 1971

__________, The development of a numerical prediction model for the planetary boundary layer. U.S. Weather Bureau, Contract Cwb-10960. Final Report, 1965

Pasquill, F., Atmospheric Diffusion, D. van Nostrand Co., London, 1962

__________, Atmospheric dispersion of pollution, Quart. J. Roy. Meteorol. Soc., Vol. 97, No. 414, 1971, pp. 369-395

__________, The estimation of the dispersion of windborne material, Meteorological Magazine, 90 (1063), 1961, pp. 33-49

Pooler, F., 1966: A tracer study of dispersion over a city, J. Air Poll. Contr. Assoc., 11 (1966), pp. 677-681

Roberts, John J., et al., An Urban Atmospheric Dispersion Model, Proceedings on Multiple-Source Urban Diffusion Models, Editor: Arthur C. Stern, U.S. Environmental Protection Agency, Air Pollution Control, Research Triangle Park, North Carolina, 1970, pp. 6-1 to 6-72

__________, An urban atmospheric dispersion model, presented at the Symposium on Multiple Source Urban Diffusion Models, Chapel Hill, North Carolina (October 1969)

Roth, P. M., et al., Development of a simulation model for estimating ground level concentrations of photochemical pollutants, Final Report, Contract CPA 70-148, Environmental Protection Agency, by Systems Applications, Inc., Beverly Hills, California, 1971

Sasaki, Y., A theoretical interpretation of anisotropically weighted smoothing on the basis of numerical variational analysis, Monthly Weather Review, 99(9), 1971, pp. 698-707

Seinfeld, J. H., Mathematical models of air quality control regions, Development in Air Quality Standards, Charles E. Merrill, Columbus, Ohio, 1970

Shieh, L. J., and P. K. Halpern, Numerical comparison of various model representations for a continuous area source. G320-3293. IBM Scientific Center, Palo Alto, California, 1971

Singer, Irving A. and Maynard E. Smith, Relation of gustiness to other meteorological parameters. J. of Met., 10(2), 1953, pp. 121-126

Sklarew, R. C., A new approach: the grid model of urban air pollution, APCA Paper 70-79, Systems, Science and Software, La Jolla, California, 1970

Slade, David (Editor), Meteorology and Atomic Energy 1968, U.S. Atomic Energy Commission, Division of Technical Information, 445 pp, 1968

Smith, M. E., and I. A. Singer, An improved method of estimating concetrations and related phenomena from a point source emission. J. of App. Met., 1966, 5, pp. 631-639

Stephens, J. J., Variational initialization with the balance equation, J. Applied Meteorology, 1970, 9(5), pp. 732-739

Stern, A. C. (ed.), Air Pollution, 2nd Edition, New York, Academic Press, 1968

__________, Proceedings of the Symposium on Multiple-Source Urban Diffusion Models, sponsored by National Air Pollution Control Administration and the North Carolina Consortium on Air Pollution, October 27-30, 1969, Chapel Hill, North Carolina, 1970

Summers, P. W., The seasonal, weekly, and daily cycles of atmospheric smoke content in central Montreal, J. Air Poll. Control Assoc., 16, 1966, pp. 432-438

Sutton, O. G., Micrometeorology, McGraw-Hill Book Co., New York, 1953

Taylor, G. I., Diffusion by continuous movements, Proc. London Math. Soc., 2(20), 1921, pp. 196-202

Wigley, T.M.L., Characteristics of Wet Plumes, presented at Conference on Air Pollution Meteorology, April 5-9, Raleigh, North Carolina, 1971

World Meteorological Organization, Urban Climates, Technical Note No. 108, WMO-No. 254. T.P. 141, Secretariat of the World Meteorological Organization, Geneva, Switzerland (1970)

Chapter IV

RESEARCH PLAN--ATMOSPHERIC CHEMISTRY AND TRANSFORMATION PROCESSES

Introduction

The Regional Air Pollution Study (RAPS) provides an unparalleled opportunity to study and improve the understanding of the chemical, physical, and biological processes that modify the concentrations of pollutants in the atmosphere. Data on meteorological parameters and pollutant concentrations will be provided not only by surface monitoring stations but throughout the mixing layer through the use of airborne laboratories. The long-term data acquisition within the RAPS will provide information on the variations of pollutant concentrations and meteorological phenomena as a function of hourly, diurnal, and seasonal changes. These data, in conjunction with an emission inventory to identify and quantify the emitted pollutants, will provide the requisite information on which to base studies of atmospheric pollutant cycles.

These studies of atmospheric pollutant cycles will cover the transport, transformation, and scavenging processes that modify the pollutant burden of the atmosphere. The characterization of these processes can provide the information necessary to verify and evaluate the capability of mathematical simulation models to predict the transport, diffusion, and concentrations of reactive pollutants over a region of interest. Atmospheric cycles of most, if not all, pollutants are poorly understood, and thus sufficient information should be generated within the RAPS to develop more satisfactory simulation models.

A more complete understanding of pollutants, reaction products, reaction intermediates, and scavenging mechanisms could define significant factors basic to the design of control strategies, abatement procedures, urban planning, and land use.

The Atmospheric Chemistry program will study the atmospheric cycles of pollutants for which air quality criteria have been issued as well as of those whose role is less well understood. Six major categories of pollutants will be considered in detail in this program: sulfur oxides, nitrogen oxides, hydrocarbons, particulate matter, photochemical oxidants, and carbon monoxide. Many of these pollutant categories are so interrelated and interreactive that a clearcut separation between them in either discussion or study is not possible or desirable. The study of particulate material--composition, size distribution, and concentration--is of particular importance as the formation of particulate material is an important scavenging mechanism for many pollutants. The studies of interreactions of hydrocarbons, nitrogen oxides, and photochemical oxidants must be integrated to characterize photochemical activity in the atmosphere. The role that precipitation plays in the removal of both gaseous and particulate material will be studied within the Atmospheric Chemistry program. The information on air quality and meteorological parameters acquired from the RAPS monitoring network will be integrated with a large number of research projects specifically directed toward acquiring information on pollutant modification within the atmosphere.

The Atmospheric Chemistry program will be initiated in late 1972 and will continue through 1978. In the latter years of the RAPS, the program will be expanded to include specific projects directed toward the concentration and cycles of the more toxic trace constituents of atmosphere, such as mercury, cadmium, beryllium, vanadium, asbestos, and polynuclear aromatic hydrocarbons. Also scheduled for the latter years would be studies concerned with atmospheric odors and stable isotope analyses of carbon monoxide and sulfates to indicate origin. Several instrument developments including sulfate, nitrate, and hydrocarbon analyzers would greatly aid the development of this chemical program.

The SO_2 Cycle

The sulfur compounds found in polluted urban atmospheres--H_2S, SO_2, and H_2SO_4, or sulfate aerosols--constitute a noxious series of compounds. Both H_2S and SO_2 have an unpleasant odor; H_2S is detectable at levels in the ppb range; SO_2 is detectable at ppm levels; both are toxic at higher concentrations. Sulfur compound aerosol particles can contribute to a reduction in visibility--a primary manifestation of smog in urban atmospheres--and have also been linked to adverse health effects.

Recognizing the adverse effects of gaseous and particulate sulfur compounds in community atmospheres, the EPA has recommended a number of air quality standards, including a limit of 0.14 ppm SO_2 over a 24-hour period to be reached no more than once a year as the maximum permissible SO_2 ambient atmospheric concentration level. Other standards deal with atmospheric particles, and some local areas have adopted H_2S regulations. Since the EPA SO_2 recommendation has been issued, increasing criticism of the regulation has been raised based on the large capital investment required to meet the emission standards. One response could be more flexible control strategy to ease the burden on emission sources and yet meet the necessity of the populace for cleaner air.

Sulfur dioxide is one of the major urban pollutant emissions. Coal and fuel oil contain significant amounts of sulfur, which is oxidized to SO_2 during combustion. Since the concentrations within the effluent streams are fairly low, it is difficult to remove the SO_2 either efficiently or economically. Another source of the SO_2 found in the atmosphere is the reaction of H_2S (from either pollutant or natural sources) and O_3.

Existing models of the transport of SO_2 account for its loss or scavenging by the application of a simple half-life factor. However, this factor cannot be accurately determined from existing data, and it appears to be strongly dependent on ambient atmospheric variables such as relative humidity (RH). Improvement in models to predict the dispersion of SO_2 will depend on determining which reaction processes actually contribute to changes in atmospheric SO_2 concentrations and the effects of atmospheric parameters. These reactions are summarized in Table IV-1. Therefore, to better define the SO_2 transformation processes, H_2S, SO_2, O_3, NO_2, NH_3, hydrocarbons (HC), relative humidity, and solar radiation should be measured regularly. Prior to the measurement program, the point sources of SO_2 emissions (such as power plants) should be located and estimates of the vegetation characteristics should be made. A program to measure the average distribution of sulfate in the aerosols as a function of particle size must be established. At the same time, an empirical correlation between the nephelometer and particulate mass loading data from the monitoring network and the aerosol particle size-distribution and total mass should be made. Several laboratory programs to establish clearly some of the reactions and inaction rates still in question are also necessary. Development of a field instrument for rapid determination of sulfate concentrations in aerosols should be encouraged.

Emission Sources of Sulfur Compounds

The largest source of H_2S is the background emissions due to the natural anerobic decay processes of organic materials. Robinson and Robbins (1968) have estimated that the worldwide background emissions will be 100×10^6 tons with an average concentration of about 0.2 ppb over land regions. However, the estimates of the total emissions of H_2S have been calculated by difference based on the additional sulfur-containing sources necessary to balance the overall sulfur cycle. The major sources of industrial H_2S emissions are primarily kraft paper mills and oil refineries. Community sources include sewage treatment plants.

Table IV-1

CHEMICAL REACTIONS OF IMPORTANCE TO THE SO_2-CYCLE

Reaction	Required Validation Data
$H_2S + O_3 \xrightarrow[\text{surface}]{} SO_2 + H_2O$	SO_2, H_2S, O_3, aerosol particle surface area
$SO_2 \xrightarrow[O_2]{h\nu} SO_3$	SO_2, $SO_4^=$ light intensity*
$NO_2 + HC + SO_2 \xrightarrow{h\nu} \text{products} + SO_3$	NO_2, reactive HC,† $SO_4^=$
$SO_2 + O_2 \xrightarrow[\text{catalyst}]{} SO_3$	RH, catalyst concn., $SO_4^=$
$SO_2 + O_2 \xrightarrow[\text{or fog}]{NH_3\text{, rain}} SO_3$	NH_3, weather, $SO_4^=$

---

* In each case, where SO_3 is a product it rapidly and irreversibly reacts with water to form H_2SO_4, which in turn forms a sulfate-containing aerosol.

† Determination of the relative importance of this reaction awaits further laboratory experimentation.

There are no paper mills in St. Louis, and the refineries in Wood River, Illinois, will probably be the only significant sources of H_2S in the area. Information from the Emissions Inventory will clarify this point. In areas not directly downwind from identified sources, the levels of H_2S should be at or below the background levels measured upwind of St. Louis.

The source of the background concentration of SO_2 (about 0.2 ppb) may be attributed entirely to the oxidation of H_2S by O_3 (Robinson and Robbins, 1968). The overwhelming majority of SO_2 emissions is related to human activities, predominantly the combustion of coal and fuel oil in power plants and heating units, smelting operations, and petroleum refining.

The SO_2 air monitoring data, collected at the CAMP station in St. Louis, showed that the annual 24-hour maximum concentration of SO_2 ranged from 0.18 to 0.24 ppm between 1962-67 (USDHEW, 1969 b). The SO_2 sources in St. Louis can be divided into two general categories: emissions from combustion of coal and fuel oil for heating purposes, in relatively small installations but spread over a large area, and point sources such as manufacturing and power plants that are superimposed on the relatively homogeneous smaller sources.

Sulfate-containing aerosols are found in stack plumes in the presence of SO_2 emissions. While there may be some low concentrations of sulfate minerals in the fuel, the majority of this sulfate is a secondary emission, formed by catalytic oxidation of SO_2 in the stack plume. While sulfate-containing aerosols from evaporated sea spray are substantial contributors to the global sulfur cycle, they should be relatively unimportant in the St. Louis area because it is so far inland.

Chemical Reactions of Importance to the SO_2-Cycle Model

The chemical reactions of importance to the development of a SO_2-cycle model can be divided into two categories: the oxidation of H_2S by O_3 and the oxidation of SO_2 to SO_3 which is then rapidly converted to H_2SO_4 by water. The latter oxidation reaction can occur via several pathways, including direct photooxidation of SO_2, oxidation of SO_2 in the presence of the NO_2-HC photooxidation system, and catalytic oxidation in stack plumes, aerosol particles, and rain or mist droplets.

Robbins (1961) found that in the presence of O_3, H_2S was oxidized to SO_2. It was later shown by Cadle and Ledford (1966) that the reaction was at least partially heterogeneous, with a rate proportional to the square root of the surface area in the reaction vessel. The reaction is so sensitive to surface catalysis that the homogeneous reaction rate could not be measured.

The complex mechanisms of the oxidation of SO_2 to SO_3 have recently been critically reviewed by Bufalini (1971). The problem may be divided into three general categories: photooxidation of SO_2; oxidation of SO_2 in the presence of light, NO_2, and HC; and catalytic oxidation by metal ions.

Numerous authors have studied the photooxidation of SO_2 in the presence of O_2. The major product was SO_3, which is rapidly hydrolyzed to H_2SO_4 (Bufalini, 1971). Experimental values of the quantum yield for SO_2 disappearance range from 0.02 to 0.22 depending on the RH and the O_2 concentration. Several authors have predicted that O_3 may be a photoproduct but this is not supported by more recent results (Sethi, 1971).

When NO_2 is added to the reaction mixture, reactions of NO and NO_2 with SO_2, SO_3, and the proposed intermediates--SO and SO_4--must all be considered (Bufalini, 1971). When reactive HC are added to the mixture and the effect of changing RH is considered, the reaction sequence becomes hopelessly complex.

Gerhard and Johnstone (1955) found no effect of added NO_2(1-2 ppm) on the photooxidation of SO_2 (10-20 ppm). In contrast, Renzetti and Doyle (1960) found increased aerosol production in a reaction mixture of 0.14 ppm SO_2 when 1 ppm NO_2 was added. Several other equally conflicting results are reviewed by Bufalini (1971). In general, enhanced conversion of SO_2 to SO_3 in the presence of NO_2 was predicted.

In the presence of reactive HC to form an SO_2-NO_2-HC system photochemical aerosols containing organic materials, H_2SO_4, and water are formed. Renzetti and Doyle's data indicate a definite increase in the loss rate of SO_2 over the SO_2-O_2 system in absence of HC and NO_2. In laboratory experiments where the aerosol formed was subjected to chemical analysis, sulfate was found in the collected aerosol when SO_2 was present at the start of the run (Endow et al., 1963).

Prager et al. (1960) and Harkins and Nicksic (1965) observed that in static smog chamber experiments with mixtures of propylene, NO, and SO_2, aerosol formation began after the conversion of NO to NO_2 was complete. Subsequent data by Wilson and Levy (1968) and from SRI laboratories confirmed these results. Therefore, NO can be ruled out as a participant in the aerosol formation mechanism. Extension of these results to the SO_2 simulation model may be difficult for two reasons.

First, in these laboratory experiments SO_2 is lost slowly before the NO_2 maximum is reached; the rate increases after the NO_2 maximum (Wilson and Levy, 1968). We suspect that reactions of SO_2 on the walls of the chambers may be of considerable importance in smog chamber experiments, especially at high RH. It is difficult to separate such losses from losses due to photochemical reactions. Second, since the environment is a dynamic system where NO, NO_2, and O_3 all coexist in substantial quantities, extension of qualitative observations from static runs, except where specific rate constants have been obtained, will be difficult.

There are indications that intermediates in the O_3-HC reactions (the O_3 is formed during the NO_2-NO-HC photooxidation sequence) also oxidize SO_2. While the SO_2-O_3 reaction is very slow (Cadle, 1956; Dunham, 1960), Harkins and Nicksic (1965) observed that oxidation of SO_2 occurred in the dark in the presence of O_3 and HC. The reactive intermediates may be alkoxyl, peroxyalkyl, or peroxyacyl radicals formed during the O_3-HC reactions. Although such reactions may be important during the day when relatively high concentrations of O_3 and free radicals are present, it should slow down after sunset and under other conditions when the O_3 concentration also decreases.

The catalytic oxidation of SO_2 by O_2 has been studied under a variety of conditions. Urone et al. (1968) found that the oxidation was very rapid in the presence of dry powdered metal oxides (aluminum, calcium, chromium, iron, lead, and vanadium). Similar results were obtained with metallic submicron particles of aluminum, iron, lead, and platinum, formed from exploding wires (Smith et al., 1969), and from particles supported on Teflon beads (Cheng, et al., 1971).

Junge and Ryan (1958) and Johnstone and Coughanowr (1958) found that manganese salts were most active in catalyzing the oxidation of SO_2 to sulfates in aqueous solution, although other salts, such as $FeCl_3$, were tested. It was found that the reaction was even faster in the presence of NH_3 (McKay, 1971). When NH_3 dissolves in the droplet, it raises the pH and neutralizes the acid formed in the droplet by the HSO_3 and SO_4 salts. The solubility of SO_2 is thereby raised, and as the concentration of SO_2 increases in the droplet, so does its rate of oxidation. It was estimated that a cloud of droplets (fog, clouds, and the like) of 10-μm radius produce 10^{-11} g $(NH_4)_2SO_4$ per hour per drop, resulting in an SO_2 lifetime of about one hour under realistic urban conditions (Robinson and Robbins, 1968).

One may observe that most SO_2 emissions do not occur under foggy conditions. However, much of the SO_2 is emitted in power plant stack plumes, where the water vapor produced during the combustion of the fuel condenses as the stack gases cool and may persist as droplets for a considerable time. Gartrell and coworkers (1963) have studied the SO_2: sulfate ratio in TVA power plant plumes using heliocopter transects at successive distances downwind of the stack. Conversion rates of SO_2 to sulfate as high as 2% per min were observed.

We have hinted at a number of interesting questions that remain unanswered about the numerous possible reaction pathways for conversion of SO_2 to sulfate. For the purposes of this Prospectus, the overriding problem is to devise experiments that will distinguish between the different processes and then to compare their relative rates. Robinson and Robbins concluded that in the daytime and at low RH (below 70%), the photochemical reactions of SO_2 and NO_2 with O_2 and HC will be of primary importance. In this case the SO_2 will be converted into essentially an H_2SO_4 aerosol. In the daytime when the RH is between 70 and 100%, the aerosols will absorb substantial quantities of water and the aerosol size distribution should shift to larger sizes. Although the photochemical mechanisms will still obtain, the rate of SO_2 conversion should be increased via the solution oxidation mechanism, catalyzed by metal salts present as nuclei or in stack plumes and aided by ammonia absorption, as discussed above. In the presence of fog or rain, the volume of condensed water greatly increases. The solution oxidation mechanism of SO_2 in the droplets should then prevail.

Removal Mechanisms for Sulfur Compounds

To complete the formulation of any simulation model for sulfur compound emissions, the mechanisms for the removal of sulfur compounds from the atmosphere must be included. These mechanisms may be divided into two general classes: (1) absorption of SO_2 by vegetation and (2) coagulation, sedimentation, impaction, and precipitation scavenging of sulfate-containing aerosol particles.

Vegetation absorbs SO_2, and damage to the plants can occur at higher SO_2 concentrations. The rate of absorption is a complex function of plant growth factors including the weather and time of day. Katz and Ledingham (1939) exposed field-grown alfalfa to SO_2 concentrations between 0.8 and 1.0 ppm. In full sun, SO_2 was rapidly absorbed; the absorption rate dropped to 20% of the full sun value when the plants were fully shaded and to below 5% at night. Chamberlain (1960) used the concept of a deposition velocity, v_g, in an attempt to quantify the loss rate. He obtained values for v_g between 0.3-0.7 cm/sec for grass. Robinson and Robbins (1968) calculated a value of 1.3 cm/sec for alfalfa using Katz's data. We suggest that a value of 1.0 cm/sec is reasonable for full sunlight, 0.2 cm/sec for full shade, and 0.05 cm/sec for the nighttime rate.

Hill (1971) has studied the absorption of SO_2 by alfalfa in a carefully controlled environmental chamber. The absorption rate was expressed as 17 $\mu\ell$ min^{-1} m^{-2} $pphm^{-1}$.

The nature and concentration of the inorganic ions in the droplets and the RH determine the size of an individual aerosol particle. The smaller particles remain suspended due to the molecular motions of the air molecules. As SO_3 or H_2SO_4 is absorbed by the aerosol droplets, the droplet grows in size because its equilibrium vapor pressure is reduced. Aerosol particles also grow by coagulation if two particles collide and stick together. Aerosols will grow by a combination of these two mechanisms, depending on atmospheric conditions, until they become so large that gravitational forces cause them to settle out. Particles also are removed by impaction onto fog and rain droplets. Under these conditions, analysis of precipitation samples as well as hi-vol samples would be necessary to account for all of the particulate matter.

The Research Program

The Emissions Inventory will locate and describe the important primary sources of H_2S and SO_2 emissions. Additional data should not be required.

Data from the ground level air quality monitoring stations will be used along with the Emissions Inventory data to calculate the incoming and outgoing flux of SO_2 in the St. Louis area. It would be useful to supplement these calculations with airborne SO_2 measurements. Barringer (Moffat, Robbins, and Barringer, 1971) has developed a technique of "correlation spectrometry" which could conveniently provide the required data. If the natural sunlight and skylight are used as the light source, the total amount of SO_2 between an aircraft-mounted monitor and the ground or above a given surface point could be determined. By flying or driving a series of transects upwind and downwind of St. Louis, the crosswind profiles of SO_2 could be obtained. From these data plus the wind velocity, the SO_2 fluxes as a function of travel time could be calculated. Such information would be of great value in the verification of the SO_2-cycle simulation model.

A field program to assess the importance of the reaction between H_2S and O_3, which yields SO_2, should be part of the Research Program. The change in the concentration of H_2S and O_3 as an air mass passes through St. Louis will be monitored by the monitoring network stations. However, a surface is required to catalyze the reaction. Atmospheric aerosols provide the largest surface in a polluted atmosphere (rather than the buildings or ground). Therefore, the total area of the surfaces present in the air mass due to the aerosols therein must be calculated. The area will depend both on the particle size distribution and the total numbers of particles at various times during the day. We recommend that a preliminary program, utilizing various types of particle counters (electric-mobility, nuclei counters, and photometric counters), be established to develop an algorithm for calculating the total surface area using the nephelometer data from the air quality network. If one assumes the size distribution determined by Junge (1963) for an "average aged continental aerosol," the problem is greatly simplified. However, it will still be necessary to relate the nephelometer readings to the total number of particles and to the aerosol size distribution. As is discussed later in this section, knowledge of the aerosol size distribution is also essential for model verification.

Currently there is no chemical model for the SO_2 cycle. One reason for this is that there is insufficient laboratory data currently available to permit one to write a reasonable reaction scheme. Although there is no doubt that appreciable quantities of SO_3 can be produced by the photooxidation of SO_2 in the presence of O_2, numerous questions remain unanswered, such as quantum yields, the effect of RH, the possible formation of O_3 in the reaction, and the effect of wavelength and light intensity on the course of the oxidation. The importance of SO_2 photooxidation in the presence of NO_2 and HC has not been adequately studied. We concur with Bufalini's recommendation (Bufalini, 1971) that the effect of the SO_2:NO_2 ratio over a wide range of concentrations should be evaluated. Better laboratory determinations of the rate constants in the oxidation mechanisms are needed, as is better data on aerosol formation and the effect of water vapor.

The effect of water vapor on the yield of photochemical aerosol is well documented. However, this effect arises in part because the aerosol size-distribution shifts towards larger sizes as RH increases. Since particles in the 0.3 to 1.0 μm range scatter light more effectively than smaller particles, and since in most experiments the yield of aerosol particles is determined by light scattering devices, the apparent yield of aerosol particles increases dramatically as RH increases. Laboratory experiments designed to separate the effect of RH on aerosol size and SO_2 conversion rates should be carried out.

Judging by the current literature and the research grants issued to laboratories at Ohio State University, the Pennsylvania State University and University of California at Riverside, these answers may be available before the start of this program. If not, programs designed to obtain them should be established as soon as possible, because they must be answered before a chemically correct simulation model can be written.

To the best of our knowledge, no one has attempted to measure the catalytic oxidation rate of SO_2 on particles produced photochemically in a mixture of NO_2, HC, and SO_2, or in the presence of trace metal salt nuclei and (or) ammonia. There is evidence that above 70% RH, most aerosols absorb water, which is accompanied by a shift in the aerosol size distribution toward larger sizes. Under these conditions, the effect of NH_3 in increasing the solubility of SO_2 in the aqueous droplet may be very important in determining the overall reaction rate. Research into this question should be encouraged.

Before the absorption of SO_2 by vegetation can be included in a simulation model, information on the type and distribution of vegetation in the St. Louis area will be needed. We recommend that the St. Louis region be divided into subregions of similar vegetation types. The surface area of vegetation within each subregion should be estimated so that this concept can be considered for inclusion in the simulation model. The appropriate data can probably be collected rather simply but the final solution of the scavenging process may require a major research effort. The goal would be to use the value for the plant surface area within the region, plus the concentration of SO_2 from the monitoring stations, to estimate the ratio of loss of SO_2 by vegetation absorption per unit area within the subregion. However, lack of precision in the various measurements may prevent conclusive results.

A complete simulation model for SO_2 will predict both the aerosol size distribution and the $SO_4^=$ concentration in the aerosol. As was discussed above, the aerosol size-distribution can be determined from data obtained by a combination of an electric-mobility and photometric counters and an Andersen cascade impactor. The distribution of $SO_4^=$ as a function of aerosol particle size can be determined by chemical analysis of the Andersen impactor samples. Since the contribution of the submicron particle (< 0.1 μm) to the total aerosol mass is small, determination of $SO_4^=$ in these particles is of less importance.

We recommend that five sites for the aerosol research program be selected, one upwind, two near the center of St. Louis, and two about 75-100 km downwind. It is recommended that periodic measurements for 1-2 weeks at regular intervals be made, starting January 1974. Chemical analyses should be performed as soon after collection as possible. These samples should also be analyzed for nitrate and perhaps HC to provide data for other research programs. Continuous measurement would mean that the number of samples for wet chemical analysis would quickly reach tremendous proportions (six samples per site per measurement) and would be accompanied by a large volume of numerical data from the particle counters. Therefore the selected time periods are suggested.

An instrument which would report automatically concentrations of either total sulfur or $SO_4^=$ content of aerosol particles for storage in the main data bank would be extremely useful for model verification. Furthermore, it would save considerable amounts of time since $SO_4^=$ determinations on hi-vol samples would not be required. Development of such an instrument should be encouraged early in the RAPS. Installation in selected monitoring stations would be highly desirable.

Studies of the stable isotope ratio (S^{32}/S^{34}) within the latter years of the RAPS could give information about the origin of atmospheric sulfur compounds. In general, the tendency is for enrichment of S^{34} in inorganic processes with enrichment of S^{32} in biological processes. The isotope ratio and changes in ratio are more or less analogous to the behavior of the stable carbon isotopes and could provide information to differentiate natural atmospheric sulfur constituents from urban emission sources.

Measurement of the deposition velocity of sulfate-containing aerosol particles will be discussed below under the heading, The Particulate Cycle.

The Photochemical Cycle--Hydrocarbons:Nitrogen Oxides:Oxidant

The most important photochemical reactions in polluted atmospheres are initiated by photodissociation of NO_2 to give NO and an oxygen atom. The ensuing chain reaction with oxygen and hydrocarbons (HC) forms products that cause many of the harmful effects related to smog, including formation of O_3, PAN, formaldehyde, and aerosols. To help regional control districts meet the Air Quality Criteria for HC and nitrogen oxides (NO_x), urban simulation models that include formation, dispersal, and losses of HC and NO_x will be written.

The major deficiency in existing meteorological models is that they do not include terms that adequately represent environmental chemistry. Before these terms can be included, the relative rates of competing reactions must be considered. One can conceive of separate simulation models for HC and NO_x, based solely on formation and loss processes. However, inclusion of the chemical reactions would make this virtually impossible since the reactions of NO and HC are intimately related. Therefore, we have chosen to discuss them in the same chapter.

The major natural source of nonmethane or generally reactive HC is vegetation, such as coniferous forests and farmland. Nitric oxide is produced by anerobic reduction of nitrates by soil bacteria. However, combustion sources such as power plants, automobiles, and space heaters emit most of the HC and NO_x found in the urban environment. These sources will be identified during the Emissions Inventory.

In the absence of HC but in the presence of sunlight, an equilibrium between NO, NO_2, and O_3 is established which can be summarized as

$$NO_2 + O_2 \overset{h\nu}{\rightleftharpoons} NO + O_3$$

When reactive HC are added, the O-atom (produced by photodissociation of NO_2) and O_3 attack on the HC is fast enough to perturb that equilibrium. During the reactions, NO is converted to NO_2, HC are consumed, and O_3, PAN, formaldehyde, and other products are formed.

The most important loss mechanism for both HC and NO_x is aerosol formation in the following sequences:

$$HC + NO_2 \xrightarrow{h\nu} \text{polymer}$$

$$NO_2 + O_3 \xrightarrow{NH_3} NH_4NO_3 \text{ in aerosols}$$

The aerosol is ultimately removed by sedimentation or precipitation. The importance of absorption of NO_2 and certain HC by plants must also be evaluated.

Validation of simulation models for HC:NO_x reactions will require a variety of data. Real time measurement of NO, NO_2, O_3, RH, and light intensity will be carried out in monitoring stations throughout the RAPS system. Additional data required by existing models include a simple breakdown of the HC distribution into two or three reactivity classes and the concentration of products such as formaldehyde, total aldehydes, and PAN. The program for determining the HC distribution should be divided into three phases. The first would involve construction of a HC classification instrument for installation in a number of monitoring stations. These instruments could be based on selective absorption of olefins and aromatics prior to detection by a total HC analyzer. The second phase would be the determination of the detailed HC distribution by gas chromatography at five sites, and use of a coupled gas chromatograph-mass spectrometer to determine organic materials including the oxygenated compounds at selected intervals during the RAPS. Formaldehyde and total aldehydes will be measured during the HC gas chromatography program. The determination of PAN by either long-path infrared or electron capture gas chromatography would not be considered a routine analysis at the concentrations anticipated in the St. Louis area. The infrared instrumentation is expensive and of limited sensitivity. The gas chromatographic method is difficult and requires very skilled technicians to perform satisfactory analysis. In either case, analysis of PAN using current state-of-the-art instrumentation would be expensive. Unless a need for PAN concentration data exists within a particular research project or program, the cost-to-benefit ratio would probably preclude routine determination of PAN in the RAPS program. Improvements in the state of the art for PAN analysis may occur during the span of the RAPS program; however, it probably would remain a research task rather than a technique suitable for routine analysis.

Sources of Nitrogen Oxides

The most plentiful nitrogen oxide, N_2O (the average background level is about 0.25 ppm) is produced by biological action of soil bacteria on nitrogen compounds. Other processes taking place in the soil are apparent sinks for N_2O.

The major natural source of nitric oxide, NO, is anerobic reduction of nitrates by soil bacteria. Robinson and Robbins (1968) estimated mean NO values of 2 ppb over land areas and 0.2 ppb over polar and ocean regions. In urban areas the pollution sources, mainly from combustion processes, give concentrations far above the background levels. For instance between 1964 and 1968 the maximum average NO concentration in St. Louis over a 5-min interval was 0.44-0.84 ppm (USDHEW, 1970a). The location and emission level of NO sources in the St. Louis area will be determined as part of the Emissions Survey. Major point sources will be power plants and industries. Area sources will include domestic heating and cooking units and automobiles. Urban area NO_x sources are typically more-or-less evenly divided between stationary and mobile sources.

Natural emissions of NO_2 appear to be unimportant. Major pollution sources will be the same as NO, but the maximum average NO_2 concentration in St. Louis over a 5-min interval ranged from 0.17 to 0.31 ppm between 1964 and 1968 (USDHEW, 1970a). The majority of the NO_2 is not from primary sources, but from oxidation of NO by either O_2 in stack plumes or by photochemical or natural O_3.

Significant concentrations of nitrate are found by chemical analysis of particulate matter collected in polluted atmospheres. However, primary sources of nitrate-containing particles have not been identified, although nitric acid is emitted in small quantities by sulfuric acid plants. The probable sources of nitrate particles will be located during the Emissions Survey. The nitrate is most likely found in secondary particles, formed by oxidation of NO_2 by O_3 to give nitric acid which is subsequently neutralized by ammonia.

Ammonia is not a nitrogen oxide, but it plays a special role in aerosol formation reactions. The major natural source is bacterial breakdown of amino acids in the soil. Since NH_3 is also a weak base, the emission level is pH dependent. Average background concentrations of NH_4^+ in aerosols are 3-10 $\mu g/m^3$, and vary considerably by locality. Combustion processes may be minor sources of ammonia in urban areas.

Reactions of Nitrogen Oxides in the Absence of Hydrocarbons

In this section we shall consider the reactions of NO_x in the absence of HC. We shall consider reactions with O_3 here, even though the O_3 is formed in significant concentrations by photolysis of NO_2 in the presence of HC.

In the troposphere, N_2O is essentially an inert compound. In the stratosphere, photodissociation can occur, but radiation of wavelengths below 2500 Å, which never penetrates the troposphere, is required (Bates and Hays, 1967). These reactions need not be considered further.

The oxidation of NO by O_2 is possible but very slow at representative NO concentrations in urban atmospheres. Reactions of NO and NO_2 with O_3 have been studied in great detail (Johnston and Crosby, 1954; Ford, et al., 1957). The reaction

$$NO + O_3 \rightarrow NO_2 + O_2$$

is the primary conversion mechanism of NO to NO_2 in both remote and urban atmospheres. The rate constant is high and in static smog chamber experiments, measurable yields of O_3 do not appear until all of the NO has been consumed.

The photolysis of NO_2 to give NO and an O-atom, which occurs during the daylight hours, is the single most important reaction occurring in polluted atmospheres. The O-atom reacts with O_2 to give O_3, which in turn oxidizes the NO to NO_2.

$$NO_2 \xrightarrow{h\nu} NO + O$$

$$O + O_2 \rightarrow O_3$$

$$O_3 + NO \rightarrow NO_2$$

In the absence of HC, an equilibrium is rapidly set up and

$$NO_2 + O_2 \overset{h\nu}{\rightleftarrows} NO + O_3$$

as long as HC are absent, little else would happen. The chain reaction which occurs in the presence of HC is discussed in a later section.

The reaction of NO_2 with O_3 is about 500 times slower than the oxidation of NO. However, this is sufficient to provide an important loss mechanism for NO_2. The equilibrium reaction of NO_3 with NO_2 is established very rapidly, but has not been studied quantitatively. Since N_2O_5 reacts rapidly with water to give nitric acid, the equilibrium is displaced to the left

$$NO_2 + NO_3 \rightleftarrows N_2O_5$$

$$N_2O_5 + H_2O \rightarrow 2HNO_3$$

Leighton, using representative values of NO_2, O_3, NO, and water in polluted atmospheres, estimated that the equilibrium concentration of HNO_3 should be 300 ppm, far higher than has been observed. The maximum rate of formation should be 9.2 ppm hr^{-1}. Obviously, equilibrium is never reached, probably because the rates are slow. However, these arguments suggest that this could be an important loss mechanism for NO_2 in polluted atmosphere (Leighton, 1961). Recent results obtained in these laboratories suggested that when these reactions take place in the presence of NH_3, a nitrate-containing aerosol was formed, provided the RH was above about 70%. Presumably the NH_3 neutralized the HNO_3, forming a soluble salt which is hydroscopic if the RH is above 70%. Such a process could be the major source of nitrate-containing aerosol in an urban environment (Robbins et al., 1959).

Other possible loss mechanisms have been considered:

$$NO + NO_2 + H_2O \rightleftarrows 2\ HNO_2$$

$$3NO_2 + H_2O \rightleftarrows 2HNO_3 + NO$$

While the equilibrium constants suggest pphm concentrations of HNO_3 in urban atmospheres, the rates of both reactions are slow and their contribution to the NO and NO_2 loss is probably small. Robbins et al. (1959) have suggested that in the presence of a NaCl-containing aerosol, these equilibria may be established more readily.

On a worldwide basis, the absorption by plants and the soil of NO_2 is significant. Robinson and Robbins (1968) assumed an average deposition velocity of 1 cm/sec, based on results by Tingey (1968) for alfalfa and oats.

In conclusion, there are two sink mechanisms for loss of nitrogen oxides in urban areas: sedimentation or rainout of nitrate-containing aerosols and absorption of NO_2 by vegetation. Obviously, competition between these pathways depends on the daily weather variations and the surface area of vegetation in the St. Louis area. Land use data should be gathered early as part of the RAPS program. It is recommended that the St. Louis area be divided into subdivisions to simplify the determination of the vegetation surface area.

Hydrocarbon Sources and Removal Processes

The major HC in the atmosphere is methane, which can be found in concentrations above 1.5 ppm. The major methane source is decomposition of vegetation. However, it is relatively unreactive when compared to other HC. The reaction with O_3 is extremely slow; reactions with O-atoms or hydroxyl radicals are at least an order of magnitude slower than reactions with other HC. Therefore, since in ppm concentrations methane is harmless to humans, we need not consider it further.

Vegetation, especially coniferous forests, sagebrush, hay fields, and so forth, emits large quantities of volatile HC, such as terpenes. Many of these compounds are strained, substituted olefins, and are relatively reactive in forming photochemical smog (in the presence of NO_2) (Went, 1960, 1966). The bluish haze often observed over such forested areas (the classical example being the Great Smoky Mountains) is attributed to photolysis of NO_2 in the presence of terpenes, which produces sizable quantities of aerosol (West, 1960, 1966; Ripperton, 1970). These emissions will be the majority of the background HC level in the RAPS.

There are many sources of HC in an urban area. Combustion of HC fuels, including coal, fuel oil, and gasoline, results in emissions of a wide variety of carbon-containing materials. The majority of the HC are in the C_1-C_{10} range, although low concentrations of polycyclic aromatics and other high molecular weight HC are present. Oxygenated species--acids, aldehydes, and alcohols--are also formed in small amounts during combustion. Major point sources will be power plants and coal-burning heavy industries. The urban area will itself be a diffuse area-source, due to heating, automobiles, and the like. Evaporative losses from gas mains (mainly CH_4), solvent evaporation, petroleum products from sources, such as gas tanks, carburator bowls, and fueling operations at service stations, will also provide significant HC and oxygenated and halogenated carbon-containing materials.

The most important removal process of HC occurs in the presence of NO_x and light by photochemical reactions. In view of the complexity of these reactions, they are considered in the next section. Ultimately, these materials are absorbed by aerosol particles, which are removed by either sedimentation or by rain or fog. Absorption losses by vegetation are also a possible sink for certain HC. For instance, ethylene in low concentrations causes extensive damage to plants (USDHEW, 1970a). The damage by other carbonaceous materials is less well understood, and the relative importance of this process compared to photochemical reactions is not known.

The Hydrocarbon-Nitrogen Oxides Reactions

The HC:NO_x photochemical reactions are important because they result in many of the manifestations of urban smog. For detailed discussion, we suggest the reviews by Leighton (1961) and Altshuller (1971).

The equilibrium between NO, NO_2, and O_3, which is established in the presence of light and O_2, is upset by HC. They react with the O-atoms and O_3 to form oxygenated photoproducts and free radicals. The products include formaldehyde, other aldehydes, PAN, and its homologues such as PBzN. For instance, olefins react more rapidly with O_3 than other HC to give aldehydes and ketones. O-atom attack on most HC is 10^3-10^4 times faster than the O_3 reaction and is less selective. In general, a chain reaction is started, which includes the following reactions

$$\mathrm{O + RH \rightarrow R\cdot + OH}$$

$$\mathrm{R\cdot\ (or\ H\cdot) + O_2 \rightarrow RO_2\cdot\ (or\ HO_2\cdot)}$$

$$\mathrm{RO_2\cdot\ (or\ HO_2) + NO \rightarrow RO\cdot\ (or\ HO\cdot) + NO_2}$$

$$\mathrm{RO_2\cdot + NO_2 \rightarrow PAN\ or\ its\ homologues}$$

$$\mathrm{RO\cdot\ (or\ \cdot OH) + R'H \rightarrow R'\cdot + ROH\ (or\ H_2O)}$$

$$\mathrm{RO\cdot\ (or\ \cdot OH) + CO \rightarrow R\cdot\ (or\ H\cdot) + CO_2}$$

When one considers a specific compound in place of RH above and considers rearrangement of the radical fragments R•, many more steps are possible. For instance, Altshuller (1965) lists 39 steps for a general olefin reaction sequence.

The kinetic equations for such a complicated reaction sequence cannot be solved without the aid of a computer. These attempts have been called simulation models. A variety of these models have been written and the published information is listed below (Seinfeld, et al., 1971).

- Highly simplified mechanism (fewer than ten reaction steps)

Friedlander and Seinfeld (1969)	general*
Eschenroeder (1969)	general
Behar (1970)	general

- Simplified mechanisms (10-25 reaction steps)

Wayne and Earnest (1969)	propylene
Behar (1970)	propylene
Seinfeld, et al. (1971)	general

- Complex mechanisms (more than 25 reactions steps)

Westberg and Cohen (1969)	isobutylene
Wayne, et al. (1971)	general

---

* Refers to HC species for which the mechanism was developed.

In the interest of computation time, it is necessary to write the simplest mechanism possible which will include the major reactions. The Systems Applications, Inc. (SAI) 14-step model (Seinfeld, et al., 1971) is given in Table IV-2, along with the monitoring data required for verification (excluding meteorology and source data). These data include solar light intensity, NO, NO_2, O_3, RH, nitrate either as gaseous HNO_3 or nitrate in the aerosol, CO, HC, aldehydes, and PAN.

Several problems are immediately obvious. PAN is formed only if $R = CH_3C{=}O$. The aldehydes include formaldehyde plus aliphatic and aromatic aldehydes, depending on the structure of R. At the minimum, formaldehyde and total aldehydes should be measured during the RAPS.

The HC in the urban atmosphere include about 125 identified compounds, excluding polycyclic aromatics such as benzanthracene. The problem is that the reactivity of these HC can vary by about a factor of 100, depending on how one defines reactivity. Table IV-3 lists one such reactivity scale, proposed by Glasson and Tuesday (1970). The values of k_9, k_{10}, k_{11}, α, β, and ζ (Table IV-2) will depend on the nature and reactivity of the HC mixture. The general problem of how to incorporate the relative reactivity of different HC mixtures into a simulation model is currently being studied in several laboratories. Loss mechanisms such as aerosol formation are not included in current simulation models, nor are the effects of RH or SO_2, but will be a necessary part of future models.

The Research Program

The most important variables, except for HC reactivity, that are listed in Table IV-2 will be determined automatically at monitoring stations throughout the RAPS. These include NO, NO_2, O_3, light intensity, RH, and CO. While total HC and CH_4 will also be automatically measured, these data will not be sufficient for future more sophisticated models. Furthermore, no provision has been made for determination of formaldehyde, total aldehydes, or PAN. These data must be collected as part of the Research Program.

The ideal simulation model would require as inputs the concentrations of the 25 to 50 more reactive HC, as determined by an analytical gas chromatograph (gc). In that case, the value of the rate constants and product yields for each HC can be determined quite accurately. Although present models are not able to handle such a mass of data, we recommend that some information about the HC distribution should be acquired early in the RAPS. The major difficulty in the gc technique is

Table IV-2

THE 14-STEP SAI MODEL

Reaction	Required Validation Data
$NO_2 + h\nu \xrightarrow{1} NO + O\bullet$	NO_2, light intensity
$O + O_2 + M \xrightarrow{2} O_3 + M$	O_3
$O_3 + NO \xrightarrow{3} NO_2 + O_2$	NO
$O_3 + 2\ NO_2 \xrightarrow[H_2O]{4} 2\ HNO_3{}^{*}$	RH, nitrate in aerosol
$NO + NO_2 \xrightarrow[H_2O]{5} 2\ HNO_2$	HNO_2
$HNO_2 + h\nu \xrightarrow{6} OH\bullet + NO$	
$CO + OH\bullet \xrightarrow[O_2]{7} CO_2 + HO_2\bullet$	CO
$HO_2\bullet + NO_2 \xrightarrow{8} HNO_2 + O_2$	
$HC + O\bullet \xrightarrow{9} \alpha\ RO_2\bullet$	Hydrocarbons
$HC + O_3 \xrightarrow{10} \beta\ RO_2\bullet + \gamma\ RCHO$	Aldehydes
$HC + OH\bullet \xrightarrow{11} \zeta\ RO_2\bullet + \epsilon\ RCHO$	
$RO_2\bullet + NO \xrightarrow{12} NO_2 + \theta\ OH\bullet$	
$RO_2\bullet + NO_2 \xrightarrow{13} PAN$	PAN
$NO_2\bullet + NO \xrightarrow{14} NO_2 + OH\bullet$	

---

* Reaction 4 is a composite of these reactions:

$$O_3 + NO_2 \xrightarrow{4a} NO_3 + O_2$$

$$NO_3 + NO_2 \xrightarrow{4b} N_2O_5$$

$$N_2O_5 + H_2O \rightarrow 2\ HNO_3$$

Therefore it is not necessary to retain NO_3 in the mechanism. The HNO_3 most likely forms the nitrate found in photochemical aerosols, probably after neutralization by NH_3.

Table IV-3

HYDROCARBON REACTIVITY IN NO PHOTOOXIDATION*

Internal olefins with two double bond substitutions	100
Cyclopentenes, internal olefins with one or no double bond substitutions	40
Cyclohexenes, tri- and tetra-alkylbenzenes, diolefins	15
Terminal olefins, dialkylbenzenes	5
Mono alkylbenzenes, C_4 and higher paraffins	2
2,2-Dimethylpropane, C_1-C_3 paraffins, benzene	0

---

* Glasson and Tuesday, 1970.

that the equipment is expensive, a highly trained person is required to operate it, and one analysis takes 1-2 hours. Furthermore, future simulation models will probably be designed for day-to-day operation and rapid predictions will be needed for daily air pollution control strategies.

At least one simulation model (Wayne, 1971) requires the division of HC into reactive and unreactive classes. Several laboratory HC classifiers have been reported. A promising instrument designed for the analysis of auto exhaust was described by Klosterman and Sigsby (1967).

A substractive scrubbing unit was coupled to a standard FID total HC analyzer. The HC were separated into three groups: olefins and acetylenes, aromatics excluding benzene, and other HC including benzene. Olefins and acetylene were selectively removed by a scrubber column containing mercuric sulfate. Alcohols, ketones, and organic acids also were removed by this column. Aromatics, excluding benzene, plus aldehydes, ethers, and esters were all removed by a palladium sulfate column. The analysis was performed by determining total HC (that is, with no scrubbing columns), then successively adding each scrubber. Automation of the device should be relatively easy. Therefore, we recommend that a fully automatic HC classifier should be developed, whose output is compatible with the central data facility.

Gc analysis of the HC would provide a complete breakdown of the HC distribution. In view of the high instrument cost and the fact that automatic instrumentation is currently not available, we recommend installation of this equipment in three research sites. One should be well upwind of St. Louis, one immediately downwind of the Wood River area, and the third about 30 km farther downwind.

Specific wet chemical techniques for formaldehyde, by the chromotropic acid method (Altshuller, et al., 1961), and total aldehydes, by the MBTH or other methods (USDHEW, 1970), are available. The chromotropic acid method is convenient and specific for formaldehyde. Methods for total aldehydes are imprecise because organic and higher molecular weight aliphatic aldehydes are not detected. We recommend that hourly average formaldehyde and total aldehyde measurements be made by the gc operator while the HC distribution program is being carried out.

It is extremely difficult to measure PAN routinely. The two available methods--long-path infrared or electron capture gc--require continuous monitoring by a highly trained technician. A small research program using one gc should be set up for occasional use during the aldehydes and HC distribution program. If the need for more extensive PAN measurements becomes more apparent, then construction of an automatic or semiautomatic electron capture gc should be considered.

A principal loss mechanism for HC is the incorporation of HC photooxidation products into the aerosol particles. Little is known about the chemical nature of these materials, but data obtained by the Bay Area Air Pollution Control District suggest that up to 50% of the aerosol mass collected on hi-vol samples is carbonaceous.* To study the nature of the carbonaceous materials, an aerosol sample, collected on a glass fiber filter, could be gradually heated in a vacuum line. The volatilized material could either be separated into its components on a gc column followed by detection by mass spectrometry, or placed directly in the inlet system of the mass spectrometer. Scheutzle (1971) has reported the development of the latter type of instrument, using a high resolution mass spectrometer and a dedicated computer. Data from such an instrument would be invaluable to the RAPS Research Program. Furthermore, the same instrument, equipped with a gc input, could be used to provide even more detailed information about the HC distribution as well as the oxygenated components in the atmosphere.

---

* This is mostly the nonvolatile portion of the aerosol.

The Particulate Cycle

To help urban areas meet the Air Quality Criteria for particulate matter, more data must be accumulated about the factors that determine the amount of particulate matter in the atmosphere at a given time. The ultimate objective of this research program will be to obtain the data necessary to develop and validate a simulation model for particulate matter. This model would, ideally, predict the particulate distribution and concentrations at some later time, given a series of inputs including the current meteorological conditions and emission levels. Both development and validation are part of this program because there are no available particulate models at this time.

Particulate matter constitutes the most visible manifestation of a polluted atmosphere. Primary stack emissions are often visible from distances of several miles away, and even quite dilute aerosols can significantly reduce the visual range. The brown color often associated with urban haze and smog may be due to preferential light scattering of certain wavelengths of light by the aerosol particles, although some authors believe it is due to absorption of light by NO_2. In addition to the visible effects of particulate matter, sedimentation of particles causes soiling of buildings, textiles, painted surfaces, and so forth. Inhalation of aerosol particles can cause a variety of respiratory diseases, partially depending on chemical composition and particle size.

In view of these detrimental health and environmental effects of particulate matter, Air Quality Criteria have been established. To achieve these Criteria, many steps must be taken. Adequate control of primary particulate emissions awaits engineering developments beyond the scope of the RAPS; however, development of simulation models covering formation and dispersal of both primary and secondary emissions will greatly aid the control strategy for air pollution. Present models, based on meteorological factors are inadequate, because secondary emissions have not been included. Validation of present and future models will depend on data accumulated in programs such as the RAPS.

The formation mechanism of particulate matter will be divided, somewhat artificially, in several categories, which include:

- Natural background aerosols--pollen, dust, and sea spray
- Primary particulate emissions--power plants, industrial emissions, and automobiles
- Secondary particles--sulfates, nitrates, photochemical products, and carbonaceous aerosols formed in the atmosphere from gaseous emissions.

Natural background aerosols contain chloride from evaporated sea spray, sulfate from oxidized natural emissions of H_2S, silicates from dust, ammonium from natural ammonia emissions, photopolymerized organic materials from natural HC emissions, such as forests, and nitrate from oxidized NO_2.

Primary particulate emissions are found in many sizes and compositions. Most obvious are the stack plumes of power plants and other industries, but emissions from domestic heating units, refuse disposal, and automobiles are also significant. The location and strength of these sources will be established during the Emissions Inventory.

Secondary particle formation can be divided into several processes which are listed in Table IV-4. Oxidation of SO_2 in the presence of either light or a catalyst, such as NH_3 or a metal ion, are fairly well understood. However, the relative importance of catalytic oxidation in stack plumes should be investigated further. The last reaction involving NO_2, O_3, and HC is extremely complex, and aerosol formation can proceed in the absence of either SO_2 or HC. Further laboratory studies of these reactions are needed to evaluate the relative rates of competing steps.

Measurements of NO, NO_2, SO_2, H_2S, O_3, and HC at the regularly reporting monitoring stations should be adequate for the model validations involving gaseous pollutants. Although the mass concentration and some information of the aerosol size-distribution can be obtained in the regular monitoring network from the particle mass monitor and the integrating nephelometer, these data will not be sufficient for model validations dealing directly with particulate concentrations. More complete knowledge of the aerosol size distribution and the concentration of sulfate, nitrate, and HC in the aerosols will be required. Currently these data observations require considerable manual sampling and analysis. As part of RAPS, the development of field instruments for rapid determination of sulfate and nitrate in atmospheric particles should be encouraged. Otherwise, collection and analysis of hi-vol samples will be necessary. A study of the average particle size distribution and the distribution of sulfate and nitrate as a function of particle size is recommended. It will probably not be necessary to monitor these variables throughout the RAPS, but representative values for the St. Louis area should be obtained.

Background Haze

The general problem of atmospheric haze formation has been recently reviewed by Germogenova, Friend, and Sacco (1970). We shall assume that the reader is familiar with this reference since we cannot hope to duplicate it here.

Table IV-4

CHEMICAL REACTIONS OF IMPORTANCE TO THE PARTICULATE CYCLE

Reactions of Known Importance	Validation Data Required
$\left.\begin{array}{l} SO_2 + h\nu \\ SO_2 + O_2 + \text{catalyst} \end{array}\right\} \xrightarrow[H_2O]{} H_2SO_4$ (aerosol)	SO_2, NO_2, HC, light intensity*
$2NO_2 + O_3 \rightarrow N_2O_5 + O_2$	NO_2, O_3, RH, NH_3†
$N_2O_5 + H_2O \rightarrow HNO_3$ (vapor)	
$HNO_3 + NH_3 \rightarrow NH_4NO_3$ (aerosol)	
NO_2 + air + HC → polymer in aerosol	NO_2, HC, RH, O_3, light intensity*
Reactions Whose Importance Must Be Assessed During RAPS	
$3NO_2 + H_2O \rightarrow NO + 2HNO_3$	NO_2, RH
NaCl (aerosol) + $2NO_2$ → $NaNO_3$ (aerosol) + NOCl	NaCl in background aerosol, NO_2

---

* Also determined as part of SO_2-Cycle Research Program.

† Also determined as part of HC:NO_x-Cycle Research Program.

Natural Background Aerosols

There is a background level of particulate matter, even in the most isolated areas. This material is largely evaporated sea spray (NaCl), sulfate ($SO_4^{=}$) from oxidation of H_2S, usually as $(NH_4)_2SO_4$ or $(NH_4)HSO_4$, nitrate (NO_3^-) from oxidation of NO_2, and organic material. Chemical analyses of particulate matter from nonurban atmospheres are summarized in Table IV-5.

Table IV-5

AVERAGE PARTICULATE CHEMICAL ANALYSES ($\mu g/m^3$)

Component	Source Area Remote*	Urban*	St. Louis
Total mass	21.0	102	168
Benzene soluble	1.1	6.7	12.8
Sulfate	2.5	10.1	
Nitrate	0.5	2.4	
Iron	0.15	1.4	
Lead	0.02	1.1	

---

* Averages for United States; Ludwig, J. H., G. B. Morgan, and T. B. McMullen, Trans. Amer. Geophys. Union, 51, 468 (1970).

These data show that benzene-soluble materials, presumed to be organic in nature, and sulfate constitute significant fractions of these aerosols. Other data show that chloride from sea spray is also an ubiquitous aerosol constituent, but its mass concentration decreases over inland areas.

The organic materials are emitted by forests, particularly coniferous forests, hay fields, and so forth. Terpenes, which constitute a large portion of the emissions, and other reactive HC are rapidly photooxidized and photopolymerized in the presence of air, light, and NO_2. Anerobic decay of vegetation produces H_2S, which is oxidized first to SO_2 by background ozone and then to sulfate. Some sulfate is traceable to evaporated sea spray. The source of nitrate is not well understood; explanations include reaction of hydroxyl radicals with NO_2 to give nitric acid, and oxidation of NO_2 to NO_3 by ozone.

The most numerous particles in the atmosphere are the Aitken particles. They are so small (0.005-0.1 μm) that they contribute little to the aerosol mass-loading and nothing to the visibility reduction, even though the background levels are as high as 10^4 particles/cm^3. They are produced during combustion and photooxidation of organic materials as well as other natural processes. The chemical composition of these particles has not been established, partly because of the difficulties in collecting them. They apparently are not important nuclei for cloud, rain, or fog condensation, but, in view of their comparatively large surface area, could serve as sites for further reactions of other chemical constituents of polluted atmospheres, such as catalytic oxidation of SO_2 and polymerization of HC derivatives.

Particles whose radius is greater than 10 μm are so large that their rate of gravitational settling is high. Consequently, they accumulate in appreciable levels only under turbulent conditions such as dust storms. These particles have significant concentrations of chloride, sulfate, and complex silicates from wind erosion of rocks and dust. Their importance on a number basis is negligible, but it can be significant on a mass basis.

Particulate Emissions Inventory

Particulate emission inventories for St. Louis have been made frequently. The data for 1963 are summarized in Table IV-6 (USDHEW, 1969). Although the emission survey recommended in this Prospectus will undoubtedly show significant changes in amounts and types of emissions, the importance of coal combustion as the major source of primary particulate matter is probably unchanged.

Since coal contains 3 to 5% sulfur by weight, SO_2 will be emitted in significant levels. HC, NO, and NO_2 are also released during combustion processes. The distribution of HC and oxygenated organic materials is determined both by the fuel and the combustion process. An estimated 330,000 tons/yr of HC were emitted in the St. Louis area in 1967-68 (USDHEW, 1971), but the average distribution of these HC according to compounds or types of compounds apparently has not been determined. All of these emissions can play a significant role in the formation of secondary particulate matter.

Table IV-6

PARTICULATE EMISSIONS INVENTORY OF ST. LOUIS
1963

Source Class	Emissions	
	Tons	Percent
Fuel combustion	86.8	58.9%
Power generation	22.4	15.2
Coal	22.4	15.2
Natural gas	0.068	--
Industrial	39.0	26.5
Coal	38.0	25.8
Domestic	19.9	13.5
Coal	18.9	12.8
Commercial and government	5.5	3.7
Coal	5.4	3.7
Refuse disposal	15.8	10.7
Motor vehicles	4.7	3.2
Industrial processes	37.5	25.4
Cement plants	3.6	2.4
Grain industry	6.7	4.5
Ferrous metals	12.4	8.3
Other	14.0	9.5
Total	147.4	100.0%

The Chemistry of Particulate Formation in the Atmosphere

In addition to the background levels of aerosols and the industrial emissions,* there will be a large contribution from aerosols produced by oxidation of SO_2, NO_2, and HC. We shall call these secondary aerosols, to be distinguished from primary particles emitted directly from pollutant sources and background aerosols. The mechanism of the formation of these secondary aerosols is very complex and poorly understood. To simplify the problem we shall include the formation of secondary aerosols in the discussion of the SO_2 cycle (sulfate-containing aerosols) and the $HC:NO_x$ cycle (nitrate- and carbon-containing aerosols). These cycles are considered in separate sections in this Prospectus. The major deficiency in the artificial separation of mechanisms is that it is more difficult for allowance to be made for the possibility of synergistic effects between the two cycles.

The size distribution of atmospheric aerosol particles is largely controlled by the rates of coagulation and sedimentation. Coagulation of Aitken particles effectively limits their numbers. Brownian movement of the particles below about 1 μm is high enough to prevent gravitational settling. For the very large particles, gravitational forces are large enough to cause significant sedimentation.

The theory of coagulation and sedimentation of aerosol particles has been reviewed in detail by Junge (1963). We refer the reader to this work for detailed references and the mathematical treatment of the problem. Although various authors seem to argue about details and variations, it has been found typically that "well-aged continental aerosols" achieve a uniform size distribution, regardless of their origin. The size distribution is apparently maintained because the competitive physical processes of nucleation, growth by absorption and coagulation, and loss of sedimentation, reach a steady state. A mathematical treatment of these processes has been developed by Junge (1963, 1965) and others.

---

* Background aerosols constitute about one-third of the total average particulate loading in urban atmospheres; a crude calculation based on a loading of 160 $\mu g/m^3$ particulate matter, spread over a 50 × 100 mi region, 1 km high, and with a residence time of one day, suggested that about 10-20% of the total particulate loading was due to primary industrial emission. Thus about half the atmospheric particle load is due to secondary particle formation processes.

In addition to coagulation and sedimentation and because the smaller particles are in continuous motion, there is a finite probability that they will collide with a surface such as the ground or a building, and stick to it. This removal process is called impaction. The sum of sedimentation plus impaction gives a net downward particle flux.

Finally, aerosols are removed by precipitation--either raindrops or fog or mist droplets. Particles become involved with droplets either by coagulation or capture. For example, the droplets fall with a terminal velocity given by the Stokes equation and as the falling droplet sweeps out a volume of air, particles in the pathway are impacted on the droplet. The higher the mass and cross-sectional area of the particles, the greater the chances of impaction. The aerosol particles are trapped in the droplet. Collection of the precipitation followed by chemical analysis shows significant concentrations of $SO_4^{=}$, NO_3^{-}, NH_4^{+} and other cations.

To determine the relative importance of these processes, the concept of deposition velocity v_g has been suggested (Junge, 1963) where

$$v_g = \frac{\text{deposition rate}}{\text{particulate concentration}} = \frac{\text{mass/area}\cdot\text{time}}{\text{mass/volume}} = \frac{\text{length}}{\text{time}}$$

In fact, v_g is the sum of three terms

$$v_g = v_p + v_s + v_i$$

where p, s, and i refer to precipitation, sedimentation, and impaction, respectively. The value of v_p can be calculated from

$$v_p = 3.16 \times 10^{-3} \frac{kh}{c}$$

where k is the concentration of a rainwater constituent in mg/ℓ, h is the mm of precipitation per year, and the concentration c of an aerosol constituent is in $\mu g/m^3$.

The dry deposition rates, v_s and v_i, can be determined by measuring the weight of either a constituent or total mass of aerosol in a known volume and the same measurements of aerosol deposited on a surface such as a filter paper or microscope slide. Esmen and Corn (1971) used a manual optical counting technique and a microscope to determine the particle flux in Pittsburgh. The deposition velocity in the absence of precipitation was measured and used to estimate the residence time of the particulate matter, which was 10^2-10^3h for submicron particles and 10-100 hr for particles in the 1-10 μm range.

The Research Program

The research program will be designed to obtain data that will be useful for validation of both a meteorological dispersion and a chemical formation model--ultimately a combined model--using state-of-the-art instruments and current knowledge of the mechanisms of aerosol formation. The necessary inputs will include

- Meteorological data
- Background aerosol levels and chemical composition
- Primary particulate source location and emission levels from both point and area sources
- Concentrations of gaseous materials important in the formation of secondary aerosols
- Chemical composition, concentrations, and size distributions of the aerosol particles.

Meteorological data will be routinely measured at all regular monitoring stations. Additional measurements should not be necessary.

Background aerosol concentrations will be available from those monitoring stations that are upwind of the source areas from the nephelometer and mass-loading data. Methods of determining the chemical composition of these aerosols are given below.

Knowledge of the location and types of emissions, including primary particles, SO_2, NO_2, and HC from point sources, such as power plants, steel mills, and refineries will be essential to any simulation model. This information will become available at the completion of the Emissions Inventory scheduled for the initial phases of the RAPS. Ideally, data giving both the particulate mass emissions and chemical composition from the point sources on a day-to-day basis would be made available while the remainder of the particulate measurement program was in progress. Alternatively, a specific sampling station could be located immediately downwind of a particular stack to obtain specific information.

As part of the Emissions Inventory, data about area sources of particles SO_2, NO_x, and HC should be obtained. Since the daily levels may vary somewhat, the information supplied by the monitoring network stations located in areas of light industry, dense housing, and major freeways will permit some assessments of changes on a day-to-day basis to be made.

For the determination of gaseous pollutants, data from the monitoring stations will be sufficient for NO, NO_2, SO_2, H_2S, O_3, and total HC. Additional determinations of ammonia and nitric acid are necessary. After completion of the short ammonia monitoring program recommended above, a limited on-going measurement program mainly in upwind rural areas should provide adequate data for ammonia. If an instrument for detection of gaseous nitric acid can be developed for the RAPS program, the data generated would be useful in the validation of a particulate simulation model which predicts the nitrate concentration in the aerosol.

Data for the particle size distribution and chemical composition of the particulate matter found throughout the St. Louis area should be obtained. These data should be measured at selected upwind locations, near primary particulate sources, and downwind where secondary particulate formation and dispersion of the stack plumes are important. As a bare minimum, particle concentrations and chemical composition including sulfate, nitrate, and HC are required. Particle concentration data can be obtained from nephelometer data, provided one assumes a refractive index and particle size distribution for the aerosol.

Determination of the chemical composition of either the entire aerosol mass or of a size-fractionated portion of it will be time-consuming and thus the sampling and analysis program must be designed for optimum utility. There are dependable methods for analysis of hi-vol samples, but since there will be at least 10 sites collecting these samples on a regular basis throughout the program, the number of samples can reach enormous proportions. If automatic instruments for sampling particulate matter and determining the nitrate and sulfate concentrations are available and dependable, they would reduce the workload considerably. More effort could then be placed on characterizing the organic constituents in the aerosol particles. Such data would be invaluable in validating models which predict the growth of aerosols in urban atmospheres.

To determine the relative importance of aerosol loss processes, the deposition velocity of total aerosol, $SO_4^=$, and NO_3^- should be measured. Millipore filters would be preweighed and used to collect aerosols. A predetermined volume of air should be pulled through one filter mounted perpendicular to the earth's surface. A second filter, parallel to the earth's surface, should be exposed to the atmosphere but no air should be pulled through it or blown over it. The total mass of material deposited on each filter can be measured and used to calculate the deposition velocity. Analysis of rain water samples for $SO_4^=$, NO_3^-, and total nonvolatiles plus simultaneous measurement of the aerosol particle concentration will provide data for calculation of the deposition velocity by precipitation.

Development of Continuous Rainfall pH Measurement and Sequential Precipitation Collection

Precipitation is considered to be an important scavenging mechanism for many of the pollutants emitted to the atmosphere by urban sources. The role that precipitation plays in removal of both gaseous pollutants and particulate material produced as reaction intermediates and products can be defined within the RAPS program. The changes in pollutant concentrations observed by the monitoring network during periods of precipitation in conjunction with a study of the density of collected precipitation at selected sites will provide the requisite data to evaluate the effectiveness of precipitation wash-out and rain-out as scavenging mechanisms.

The instrumentation for obtaining measurements concerning the chemical properties of precipitation is not available, apparently, as an off-the-shelf item. Most components to perform the measurements are available, however, and the design of appropriate instrumentation for automated pH measurements using commercially available components should be the first priority of this research project. A tipping bucket rain gage could be the basic component in which the discarded precipitation is passed through a flow-through pH measurement cell. The measurement cell must retain enough precipitation to keep the pH electrode moist during the extended periods without precipitation (one week to ten days in St. Louis). If the volume of the measurement cell is minimal, the volume of precipitation in each bucket increment is adequate to rinse the cell free of the retained volume of the preceding increment.

A separate tipping bucket rain gage should be used to collect sequential samples for further chemical analysis. A close fitting cover activated by a precipitation sensor would protect the gage from dust fall during dry intervals. The cover must be activated to cover the gage inlet after precipitation has ceased. A rotary table containing sample vessels could be used to collect samples for further analysis on a periodic basis. Ion concentrations of the precipitation could be determined through the use of specific ion electrodes. This is a rapid, simple method of analysis that can yield valuable information about the chemical constituents present in the precipitation. Specific ion electrodes in conjunction with standard laboratory pH instrumentation can determine the ion concentration of sodium, potassium, calcium, magnesium, ammonium, nitrate, and chloride. Many other ion concentrations can be measured using this technique, but the above would be of particular interest in the RAPS program. Specific ion electrodes for the determination of sulfate ion are not currently available, but may be developed during the span of RAPS. The ratios of sodium, potassium, calcium and magnesium ion concentration can be used to determine if sea salt is present as droplet nuclei. The volume of collected precipitation should be sufficient to permit other techniques of analysis, such as for sulfate concentration, to be employed as required by the individual research programs. The measurement of pH should be made at the collection site to obviate errors due to absorption of atmospheric CO_2. The collected samples for subsequent analysis should be covered after collection to prevent contamination by dust. The collected precipitation for chemical analysis cannot be the discard from pH measurements as the sample will be contaminated by leaching of the electrolyte through the glass electrode of the pH measurement.

Precipitation measurements would not be required at all monitoring stations, but instrumentation should be installed at selected stations within the network. Currently, the Illinois State Water Survey is operating and maintaining an extensive precipitation measurement network in the area of St. Louis. Liaison between RAPS and the Illinois State Water Survey should be established to obtain and integrate the precipitation collection data into the control data archive of RAPS.

Carbon Monoxide Cycle

Carbon monoxide (CO) is found in the highest concentration of any pollutant gas in the urban environment. It is colorless, odorless, and toxic to man at higher concentrations. At low concentrations, both visual and cardiovascular effects have been noted on test subjects. Comparison of background concentrations of CO (up to 0.6 ppm) and urban concentrations (60 ppm and higher) show that the greater amount of CO in the urban environment can be associated with human activity. In view of the adverse health effects of long time exposure to 50 to 100 ppm concentrations of CO, Air Quality Criteria for CO have been issued.* To help meet these Air Quality Criteria, meteorological models has been developed that successfully model the transport of CO throughout the urban area. It is the purpose of this research program to collect more complete information than has been heretofore available for validation of these models. Furthermore, existing diffusion models assume that CO is unreactive in the environment. Recent chemical models for the HC:NO_x reactions and smog chamber experiments indicate that CO is not completely unreactive. Refinements in current CO models may require an understanding of the chemistry of CO in a polluted atmosphere. The chemical reactions of importance will be discussed. In addition, active biospheric scavenging processes and possible sources in the biosphere have been noted for CO.

---

* An average of 9 ppm over an 8-hr period or 35 ppm over a 1-hr period should not be exceeded more than once a year.

The background sources of CO include marine plants and invertebrates found only in ocean regions. Biological sources on land areas have been few but they cannot be entirely ruled out on the basis of current incidence. The oxidation of background levels of methane by hydroxyl radicals has been suggested as another important atmospheric CO source. However, these sources will be of negligible importance when compared to emission sources, primarily automobiles, in an urban area.

In 1968, the automobile accounted for 77% and industrial operations contributed about 20% of the total CO emissions in St. Louis (USDHEW, 1970B). All are due to incomplete combustion of HC fuels.

In a polluted atmosphere where NO_2 photolysis results in a significant concentration of hydroxyl radicals, a chain reaction is established:

$$OH + CO \rightarrow H + CO_2$$

$$H + O_2 \rightarrow HO_2$$

$$HO_2 + NO \rightarrow OH + NO_2$$

One molecule of NO is oxidized to NO_2 and one molecule of CO is oxidized to CO_2. While this reaction is slow compared to other potential atmospheric reactions, it is of importance to the overall HC:NO_x reaction sequence.

Recent data have shown that the soil is an important sink for CO. Of 200 fungi, yeasts, and bacteria tested, 16 species of fungi absorbed CO at concentrations of less than 100 ppm. The rate of absorption will be competitive with the losses by reaction with hydroxyl radicals.

Simulation models for CO have been developed that predict accurately the distribution of CO in localized regions, such as city streets. There are probably adequate data available now to validate these models. However, data for predicting the mass flux of CO at longer distances downwind of the sources are lacking. Such data will become available during the RAPS from the regular monitoring system and airborne measurements proposed in this Prospectus. In treating these data, the important question will be: Are loss mechanisms--chemical reactions and soil absorption--fast enough so that they must be considered as a part of CO simulation modeling?

A program designed to identify the sources of atmospheric CO upwind and downwind of the St. Louis area, using the stable isotope ratio technique, is also recommended.

Source of CO

The sources of CO can be divided into natural emissions and emissions associated with human activity. The latter sources are by far the most important in the RAPS.

Major natural sources of CO have been found which include marine plants and invertebrates (USDHEW, 1970b; Jaffe, 1968). The majority of these sources occur only in ocean areas. McConnell, McElroy, and Wofsy (1971) have suggested that a major source of atmospheric CO is oxidation of methane (CH_4) by hydroxyl radicals (OH). Swamps, rice paddies, and other areas where biological materials decompose are major sources of methane on a global basis. The following reaction sequence has been suggested (McConnell, et al., 1971; Levy, 1971):

$$NO_2 + h\nu \rightarrow NO + O$$

$$O + O_2 + M \rightarrow O_3 + M$$

$$O_3 \xrightarrow{h\nu} O(^1D) \xrightarrow{H_2O} 2\ OH$$

$$CH_4 + OH \rightarrow CH_3 + OH$$

$$CH_3 + O_2 \rightarrow CH_3O_2$$

$$CH_3O_2 + NO \rightarrow CH_3O + NO$$

$$CH_3\ O + O_2 \rightarrow CH_2O + HO_2$$

$$HO_2 + NO \rightarrow HO + NO_2$$

$$CO + OH \rightarrow CO_2 + H$$

$$H + O_2 + M \rightarrow HO_2 + M$$

$$CH_2O + h\nu \rightarrow \begin{cases} CH{=}O + H \\ CO + H_2 \end{cases}$$

Background concentrations of NO and methane were used to calculate a lifetime for CO of 0.3 yr.

The background levels of CO from these sources range from 0.02 to 0.6 μpm; the sources apparently depend heavily on the local meteorology and the proximity to urban sources. An average of 0.2 to 0.2 ppm is a reasonable concentration to be expected upwind of the St. Louis area. Data taken at CAMP sites showed that the CO levels in St. Louis will be more than an order of magnitude higher. Between 1962 and 1967, the geometrical mean concentration was 5.3 ppm and the maximum 5-min average concentration ranged from 45 to 68 ppm. These data show that on most days the background CO is unimportant compared with CO from pollution sources in St. Louis.

Stevens, Krout, and Walling (1971) have attempted to determine the source of atmospheric CO by measuring the $C^{13}:C^{12}$ ratio of CO from various automotive, urban, and nonurban sources. The $C^{13}:C^{12}$ ratio from automobile exhaust is quite different from that measured in rural areas, suggesting that anthropogenic sources are not of primary importance on a global basis. However, natural CO sources such as ocean water, kelp floats, and marine invertebrates also did not produce CO having the same isotope ratio of either automobile exhaust or rural area samples. They concluded that there is another, as yet unidentified, source of atmospheric CO. It is possible that reaction sequences such as CH_4 plus OH, which ultimately produce CO as an oxidation product, are the additional CO sources (Levy, 1971).

In St. Louis (in 1968), an estimated 77% of the CO was from transportation sources--automobile, trucks, buses--and 20% was from industrial sources. In other cities, the contribution from transportation sources is much higher, with an average of 94% in Los Angeles. Therefore, the major CO sources will be city streets, freeways, and the like, which are represented as line sources in simulation models. Some industrial point sources will be found during the RAPS Emissions Inventory. Petroleum refinery catalytic cracking units can be significant industrial sources.

The emissions of CO result from incomplete combustion of HC fuels. Recent CO emission limits of 39.0 g/mi for cars produced between 1972-74 should cause a gradual reduction of CO emission as older uncontrolled cars are replaced. The effects of these changes should be measurable as the RAPS progresses.

Important Chemical Reactions of CO

Oxidation of CO by various gaseous species in the atmosphere can occur. Such reactions include the following:

Reaction	Activation Energy (kcal)
$CO + O_2 \rightarrow CO_2 + O$	51
$CO + H_2O \rightarrow CO_2 + H_2$	56
$CO + O_3 \rightarrow CO_2 + O_2$	20
$CO + NO_2 \rightarrow CO_2 + NO$	28

In view of the high activation energies and the low concentrations of O_3 and NO_2 in the environment, none of these reactions can be of importance to the chemistry of polluted atmospheres. Kummler et al. have suggested that $O(^1D)$, $O_2(^1\Sigma g)$, and $O_2(^1\Delta g)$ at realistic atmospheric levels cannot oxidize CO directly (Kummler, et al., 1971).

In many simulation models CO has been assumed to be unreactive, especially when only short time periods are considered. Many laboratories used CO as a tracer in smog chamber experiments to follow the dilution of the chamber contents. Recent results of Westberg, Cohen, and Wilson have shown that 100 ppm of CO accelerated the formation of both NO_2 and O_3 when isobutylene and NO were photolyzed in a smog chamber (Westberg, et al., 1971). It was suggested that a chain reaction involving CO, NO, NO_2, and OH-radicals was established which increased the rate of oxidation of NO to NO_2 (McConnell et al., 1971).

$$OH + CO \rightarrow CO_2 + H$$

$$H + O_2 + M \rightarrow HO_2 + M$$

$$NO + HO_2 \rightarrow OH + NO_2$$

The result of the above mechanism is a gradual conversion of CO to CO_2 in the urban atmosphere. Hydroxyl radicals are thought to be an important chain-carrying intermediate in the HC:NO_x photochemical reactions. The concentration of OH and therefore the rate of conversion of CO will also depend on the HC, NO, and NO_2 concentration in the atmosphere. The HC:NO_x simulation models could generate an estimated steady-state concentration of OH and HO_2 radicals. These numbers could be used to estimate the rate of oxidation of CO to CO_2.

CO Sinks

A major CO sink in the biosphere appears to be the soil. Inman, Ingersoll, and Levy (1971) found that soils can rapidly absorb CO at concentrations of 100 ppm or less. Hill (1971) and Inman and Ingersoll (1971) showed that plants did not absorb measurable quantities of CO. About 200 species and strains of bacteria, fungi, and yeasts from several soil samples were isolated and cultured. Of these, 16 cultures, all fungi, were active in CO uptake. Rates of CO uptake for various soil samples ranged between 2.2 and 17 mg/hr per m^2. The major deficiency with these studies is that absorption rates at less than 5 to 10 ppm CO were not determined. Further studies are currently in progress at SRI. As far as we know, there are no other biological sinks of significance in the biosphere.

The major question is which of the two possible CO sinks--chemical reaction or biological absorption--is of greater importance in the urban environment. Using Levy's (1971) estimate of the background concentrations of OH (4×10^8 molecules/cm^3), an average CO concentration of 5 ppm, and a rate of 1.5×10^{-13} cm^3 $molecules^{-1}$ sec^{-1} for the reaction of OH and CO, we calculate a consumption rate of 7.5×10^9 molecules cm^{-3} $molecules^{-1}$ sec^{-1}.* Using Inman's data (1971) for soil absorption, we chose a representative absorption rate of 10 mg m^{-2} hr^{-1} or 6×10^{12} molecules

---

* Note that the OH concentration will be higher in polluted environments. Therefore, the rate of conversion of CO is a minimum value.

cm^{-2} sec^{-1}. The height of the inversion layer ranges from 0.5 to 5 km. At the same time, the boundary layer above the soil is about 1 cm, and about 6×10^{12} molecules sec^{-1} will be absorbed. Choosing 2 km as a representative value, for every square centimeter of soil available for the chemical reaction, about 2×10^{14} molecules sec^{-1} will be removed by the chemical mechanism. In view of the assumptions that have been made, we conclude that both chemical and biological sinks are of comparable importance.

The CO Research Program

Johnson et al. (1971) have developed an urban diffusion model for CO that predicts fairly accurately the concentrations of CO in a downtown urban area. Required inputs are the traffic patterns, wind speed and direction, the mixing depth, and the vertical diffusion rate. Validation data were collected in downtown St. Louis and San Jose, California. It was found that the CO concentration varied as much as 10 ppm from one side of the street to the other. A street-canyon submodel was developed to predict the effect of wind on concentrations at different locations in the street.

The model was reasonably successful in predicting the CO concentrations, and the program could be used either in a synoptic mode, in which CO concentrations at downwind receptor stations could be calculated from traffic and meteorological data, or in a climatological mode, in which concentration frequency distributions could be calculated based on long-term traffic and meteorological data sequences.

The overriding question which remains unanswered is whether or not the model will correctly predict the mass flux across a vertical plane downwind of the urban area. Given inputs such as the emission levels at the sources plus the wind direction and velocity, the downwind pollutant concentrations as a function of height can be predicted by the program. The mass flux can be calculated from these data. To determine the effect of chemical and biological CO sinks on the overall dispersal of pollutants, the mass flux should also be determined from actual measurements made in St. Louis during the RAPS. For these calculations, wind trajectory and CO concentration data from the regular monitoring stations should be supplemented with airborne measurements. The mixing height should be located and the CO concentrations at several altitudes at and below the inversion layer should be measured. From these data, the actual mass flux through a selected vertical downwind surface can be calculated. If the measured flux is much less than the values predicted by the model, then one must conclude that either the chemical or biological sink (or both) is removing significant amounts of CO.

This program should also include supplementary CO isotope ratio studies of samples collected upwind of St. Louis, in the downtown area, and at sites as far as 200 km downwind. Samples should be collected while the mass flux experiments are in progress. These data should show a dramatic change in the isotope ratio between upwind and downwind sites in the city. It is more difficult to predict the ratio 100 to 200 km downwind, where possible CO sinks and biological sources may affect the ratio.

The Chemical Research Program Schedule

Several research programs have been recommended in the preceding discussion, which are designed to provide supplementary but vital information for verification of simulation models. The Class A and B stations will provide complete data for meteorological models and most of the chemical data, including the visibility reduction and the concentrations of NO, NO_2, H_2S, SO_2, O_3, CH_4, and nonmethane hydrocarbons. Additional information will be required for each model that cannot be obtained with completely automated instruments because such instruments are not available. These programs are summarized in Table IV-7. In the discussion that follows, we shall consider the nature and justifications for each program relative to model development prospects. It is also likely that there will be other applications for data resulting from the chemical research program which cannot be anticipated.

A total of up to 15 of the Class A and B stations will be designated as research stations. An upwind station should be selected far enough upwind to permit reliable measurements of background pollutant concentrations. Several stations will be located near St. Louis. At least one should be just downwind of the Wood River area to measure refinery and other industrial pollutants close to their sources. Others will be perhaps 10 miles west and would be in the path of emissions from the city itself (that is, from automobiles, space heaters, and the like), while additional stations would be located at least 20 and 40 miles downwind. These stations would measure the components of the St. Louis atmosphere after several hours of transport--sufficient time for photochemical reactions to take place. The sites selected must be reasonably near good roads to minimize travel time but must not be near major pollutant sources. Atmospheric components to be measured at the sites are discussed below. It will be necessary to perform some analyses at the central laboratory facility. Complicated analyses will be sent to Durham to avoid duplication of expensive facilities.

Table IV-7

THE CHEMICAL RESEARCH PROGRAM

Program Title	Justification	Technique	Start Time	Number of Sites
Particulate cycle				
Total NO_3^- and $SO_4^=$	Aerosol is principal sink for NO_x and SO_2	Wet chemistry Automatic instruments	January 1974 July 1975	15 15 or more
Determine aerosol size distribution	Aerosol models will predict size distribution under various meteorological conditions	Electric mobility, aerosol particle counter, Andersen impactor	January 1974	5
Determine distribution of NO_3^- and $SO_4^=$ in aerosol	Concentration of NH_4NO_3 and NH_4HSO_4 determines aerosol particle size	Andersen impactor	January 1974	5
Importance of NO_2-Cl^- reactions in aerosol	Evaluate importance of this NO_2 scavenging mechanism	Wet chemistry	January 1975	5
Characterization of organic matter in aerosols	Aerosol important hydrocarbon sink; nature of organic material is not known	Gc-mass spectrometer	July 1975	1
Hydrocarbon programs				
Gc-HC analysis	Provides supplementary HC-distribution data for total HC measurements	Gc Automatic HC classifier	January 1974 July 1975	5 15
Gc-mass spectrometer HC analysis	Supplements Gc program; provides best available peak identification	Gc-mass spectrometer	July 1975	1
Formaldehyde-total aldehydes monitoring	These are products predicted in current HC:NO_x simulation models	Wet chemistry	January 1974	5
PAN (optional)	Another predicted product	Electron-capture Gc	January 1975	5
NH_3 Monitoring Program				
Phase I - determine need Phase II - routine program	NH_3 neutralizes HNO_3 and H_2SO_4, accelerating aerosol formation	(1) Wet chemistry (2) Modified NO detectors	July 1973 July 1974	5 ?
CO Monitoring (supplementary)				
Mass flux	Verify existing models by providing height profiles of CO	Airborne CO monitor	1974	1
Isotope ratio experiments	Provides experimental evidence for CO sources	Mass spectroscopy	1974	1

Aerosol Research Program

Current evidence suggests that aerosol formation is the primary loss mechanism for SO_2, HC, and NO_x. Automatic instrumentation for monitoring these aerosol components is currently not available. However, these data will be essential for model verification so that hourly concentrations of NO_3^-, $SO_4^=$, and the mass balance can be determined and compared to values calculated in the model.

Sulfate and Nitrate Compounds--It is recommended that the EPA should push for the development and testing of an instrument (or instruments) that would automatically monitor NO_3^- and $SO_4^=$ in aerosols. The output of the instrument should be compatible with the RAPS central data collection facility. Such a device should result in more data with considerable long-term savings. An outside contract would probably be the most efficient way to procure these devices. This instrument need has long been recognized and there is real doubt that current technology can conceive and produce instruments of these capabilities.

Since automatic instruments are currently not available, wet chemical analyses of hi-vol samples will be necessary. Hi-vol samples will be collected at all Class A and B stations on a semiweekly schedule. Although chemical analyses for NO_3 and $SO_4^=$, as well as NH_4^+, Pb, and other metals and HC would be desirable, the cost of routinely measuring these components is high relative to the knowledge gained. It is our recommendation that routine analyses for NO_3^- and $SO_4^=$ in samples from 10 to 15 carefully selected sites would provide adequate data and could be performed by one analyst in St. Louis. As part of the Ammonia Monitoring Program, some NH_4^+ analyses will be required. This program should begin in January 1974 or whenever the monitoring stations are put into operation. It may be possible to analyze samples collected less frequently with no significant loss in data. Such decisions can be made early in the program on the basis of the observed variability. Analyses for trace metals should be done only after a need for such data has been developed.

Organic Compound Aerosols--It would be desirable to study the carbon-containing fraction of the collected aerosol. Several laboratories have measured the infrared spectra of collected aerosols, but the technique was laborious and not very successful. We recommend that a gas chromatograph-mass spectrometer (gc-mass spec) instrument with a slave computer would provide much useful information. For aerosol analysis, a temperature-programmed pyrolysis input to the gc is recommended. The gases driven off by either evaporation or pyrolysis would be detected in the mass spectrometer and identified by the computer. It will be necessary to hire a professional to operate the instrument and to develop techniques for analyzing the samples. The cost of the instrument will be high (about $80,000), but the instrument also will be used for the analysis of gaseous hydrocarbons and oxygenated photoproducts discussed in the HC:NO_x cycle section. The program should start in July 1975.

Particle Size Distributions--Future aerosol simulation models should predict both the aerosol size distribution and the distribution of chemical species (NO_3^- and $SO_4^=$) as a function of particle size. Knowledge of the size distribution is necessary to predict the total aerosol mass, the visibility reduction, and the aerosol surface area. The last datum will be a necessary input to the $H_2S + O_3$ reaction sequence, which is a heterogeneous reaction requiring a surface as a catalyst. The aerosol size distribution research program should be operational by January 1974.

To cover the particle size range of 0.005 to 5 μm, a number of devices are necessary. Electric mobility counters are expensive but provide some idea of the size distribution between 0.005 to 0.1 μm. The condensation nuclei counter, available in both manual and automatic versions, will give the total number of particles in that size region but no information as to their size distribution. However, the cost is a fraction of the electric mobility counter and it is simpler to operate.

Particles in the 0.3 to 2-μm range cause the greatest light scattering or visibility reduction. Larger particles are also effective but the number of such particles is much lower so in effect they make little contribution. Commercial photometric particle counters are available, which are well suited to automatic operation and digital data output. Simply stated, when a particle passes through the sensing region, a pulse of light scattered by the particle is detected by a photomultiplier tube. The electronic logic system within the counter registers the pulse in a channel whose boundaries represent a size range. Counts are averaged over a preselected time interval, then read out to a digital printer such as a teletype. The system could easily be adapted to the RAPS central data storage facility. The averaging time should be between 10 and 30 min.

Andersen impactors have been used for many years to fractionate and collect the larger aerosol particles. Chemical analysis of each fraction provides information on the distribution of important solutes in the aerosols. Serious drawbacks to these devices are the time required to collect a sample large enough for analysis (6-24 hrs) and the number of samples collected per run (six). Since automatic collection and handling are impossible and the number of samples from five sites will be 30, the number of runs must be limited. We recommend that the five sites should be operated simultaneously every other day for two weeks. On these days, the other particle counters should be operated simultaneously to maximize the information collected. On alternate days, the samples will be collected and weighed and the sampler cleaned and prepared for the next day's operation. At the end of the period, the samples should be carefully labeled, packed, and shipped to EPA, Durham, for analysis.

During these studies the integrating nephelometer, located at all Class A and B stations, will be operating. After particle size distribution data and the NO_3^- and $SO_4^=$ composition have been collected for about 6 to 9 months, attempts to correlate nephelometer readings with HC, NO_x, and SO_2 concentrations, relative humidity, and meteorology should be made. Future particulate models will predict aerosol size distributions based on these inputs. If correlations between nephelometer readings and the measured aerosol size distributions could be established, they would greatly aid future model verification studies.

Chloride in Aerosols--As part of the RAPS, the importance of the reaction between Cl^- in aerosol particles and NO_2 should be evaluated, although we suspect that, since St. Louis is very far inland, there will be little Cl^- from sea spray in the background aerosols.* However, Cl^- could be provided by other sources and this mechanism could be an important NO_2 scavenging process. Therefore, a limited number of Cl^- determinations in aerosols collected on hi-vol samplers should be made. If the Cl^- concentrations are high, more detailed studies will be required and an expanded program could be planned. Initial studies could be made at convenient times after January 1975.

The SO_2 Flux Measurement

Since the SO_2 flux through the St. Louis area could be calculated by simulation models, it would be useful to measure the flux independently to verify the model. The correlation spectrometer designed by Barringer

---

* Junge (1963) indicates that the Cl^- content of precipitation in the central midwestern area is about 0.2 mg/1. This figure is about 10% of the $SO_4^=$ content, 20% of the NO_3^- concentration, and greater than the NH_4^+ concentration.

(Moffat, Robbins, and Barringer, 1971) should be used to measure the SO_2 altitude profiles throughout the St. Louis area. Airborne instruments are currently available. The SO_2 profiles and the wind velocity measurements from Class A and B stations will be inputs to a computer program which calculates the SO_2 flux. These measurements should be made several times a year, beginning in 1976.

It may be possible to gather similar data for NO_2, since it also can be detected by correlation spectroscopy. By 1976, instruments for CO, NH_3, and other gases that have distinctive infrared absorption spectra may be available. If so, then programs for measuring their fluxes should be considered at that time.

The HC:NO_x Research Program

The HC:NO_x research program can be divided into two distinct categories: the distribution of HC in the urban atmosphere and the concentration of the principal photoproducts of the HC photooxidation reaction. The latter compounds include formaldehyde, total aldehydes, and peroxyacetyl nitrate (PAN), whose concentrations are specifically predicted in current HC:NO_x simulation models.

HC Distribution--The reactivity range of the various hydrocarbons found in an urban atmosphere is about two orders of magnitude. For satisfactory modeling, some estimate of the reactivity of the HC mixture will be necessary. We recommend that determination of the HC distribution should be broken down by the use of three types of instruments: (1) the HC classifier, (2) an HC gas chromatograph (gc) and (3) a gc-mass spectrometer.

The HC classifier would be a new instrument design based on a total HC analyzer. By using a series of programmed switching valves, several absorption tubes can be selectively switched into the sample line for the analyzer. Klosterman and Sigsby (1967) used scrubbers containing mercuric sulfate to remove olefins, acetylene, alcohols, ketones, and organic acids and scrubbers containing palladium sulfate to remove aromatics (excluding benzene) and aldehydes. The analysis was performed by determining total HC, then successively adding each subtractive column to the inlet stream. An automatic instrument could be built whose switching system would be triggered by the interrogation signal from the RAPS data system and whose output would be compatible with the RAPS data system. Five minute interrogation intervals would allow time for the instrument to equilibrate before readout to the data-handling system. Since it would be relatively simple, inexpensive, and entirely automatic, we recommend that at least 15 instruments be procured. It might be more efficient to award an outside contract to develop, test, and deliver the instruments. They would be located at selected upwind, downtown, and downwind sites. Since operation should commence by January 1975, construction should begin before January 1974.

Although the prospective HC hydrocarbon classifier should be simple and automatic, it would not provide a complete breakdown of the HC composition. Satisfactory gc techniques using freeze-out loops and temperature-programmed capillary columns have been developed and used for similar experiments. However, the instruments are expensive and require an experienced operator. Therefore, a maximum of five research sites should be established. It will be necessary to have a technician present while the gc is being operated, but he will be available for other tasks at the same time.

The gc output should be fed directly into a digital integrator, which would record the retention time and area for each peak. If suitable instrumentation could be purchased, it would be best to have a single integrator located at the central laboratory in St. Louis which would monitor all five instruments simultaneously. Instead of requiring five integrators at $6,000 each, one $30,000 computer could be purchased which would be much more flexible and save time in later data workup. The biggest problem will be the identification of the gc peaks. The mixtures that can be anticipated immediately downwind of the city are so complex that completely automatic peak identification may be impossible. Data workup procedures must be developed during the test and break-in periods of the study. Therefore, acquisition of the required instrumentation should begin in early 1974. The system should be operational in January 1975.

The technicians who will carry out the aerosol particle collection experiments would also operate the gc apparatus. A staggered monitoring program would minimize the work load.

The most satisfactory way to determine the HC distribution is also the most complicated: gc-mass spectroscopy. In theory, complete identification of all compounds, including oxygenates, is possible. In many cases, compounds that elute under the same peak can be identified separately.

Due to equipment, personnel, and operating costs, only one gc-mass spectrometer should be purchased and installed at the central laboratory facility. Samples would be collected in evacuated glass or stainless steel bottles at the five research sites and brought to the central facility for analysis. Coordination with the gc operation at the research sites is desirable. In this way, scheduling conflicts with the aerosol identification program can be avoided.

Formaldehyde, Total Aldehydes, and PAN--Formaldehyde, other aldehydes, and PAN are all photoproducts whose concentrations as a function of time are predicted in current simulation models. Therefore, a program to monitor these materials is recommended for the RAPS to begin January 1974 at the five research sites. The major difficulty is that automatic monitoring instruments are not available.

The chromotropic acid method for formaldehyde is specific, simple, and thoroughly tested (USDHEW, 1970a). It is, however, a wet chemical method. The sample is bubbled through a solution of chromotropic acid in concentrated sulfuric acid. The collected sample solution is stable for several days. Therefore, hourly samples from all sites could be collected, brought to the central laboratory, and analyzed the next day. A colorimeter, such as a Bausch and Lomb Spectronic 20, is required for the analysis.

There are several methods available for determining total aldehydes (USDHEW, 1970a). The 3-methyl-2-benzothiazole hydrazone (MBTH) method (Altshuller et al., 1961) has been recommended by EPA as most suitable for routine analysis (USDHEW, 1970a), even though aromatic aldehydes are not quantitatively determined. Alternative methods, such as trapping in bisulfite, are known. Two analytical methods have been used. The uncomplexed bisulfite can be titrated with iodine. Alternatively, the bisulfite complex can be decomposed by acid and the released aldehydes reacted with 2,4-dinitrophenyl hydrazine. However, the resulting phenyl hydrazone derivatives have different absorption curves, complicating the colorimetric analysis. Detection of higher aldehydes is apparently limited by their solubility in water. On the other hand, it was recently shown in these laboratories that benzaldehyde was quantitatively trapped, suggesting that the method may be more suitable than the MBTH method if it could be shown that higher aromatic aldehydes can also be quantitatively trapped. Therefore, we suggest that whereas the MBTH method is commonly accepted, it should not be used without careful consideration early in the RAPS program.

The well-known eye irritant peroxyacetyl nitrate (PAN) is also a commonly predicted photoproduct in HC:NO_x simulation models. Its concentration in Los Angeles rarely exceeds 0.05 ppm (USDHEW, 1970a). We expect that the PAN concentrations in St. Louis should be even lower since that atmosphere contains a higher concentration of chemically reducing species.

Two methods of analysis are used for PAN detection: long path infrared and electron capture gas chromatography. Both are difficult methods. The long path infrared method can be ruled out immediately because the equipment costs are high, it requires a skilled operator, and, most important, it is of limited sensitivity (0.05 ppm). Electron capture gas chromatography has a sensitivity to PAN in the ppb range and also a low sensitivity to interfering compounds, such as hydrocarbons. Current manual techniques are satisfactory, provided daily calibrations with ethyl nitrate are performed. An experimental automatic detector has been developed (Stevens, 1969).

In view of the very low concentrations expected in the St. Louis area and the difficulties in using the gc method on a routine basis, we believe that the anticipated usefulness of the data does not warrant a PAN measurement program for the RAPS research program at this time. If later considerations suggest that PAN measurements would be useful, then an automatic instrument should be developed with an output that is compatible with the central data storage system.

NH_3 Monitoring Program--This program is designed to assess the importance of NH_3 in the formation of particulate matter. Initial experiments should determine the background concentration of NH_3 upwind of the St. Louis region. The change in concentration of gaseous NH_3 as an air parcel passes through the urban area should be measured. If there are no NH_3 sources in the St. Louis area, then the total mass of NH_3 should not change. However, as aerosol is formed, the NH_3 will be absorbed and partially neutralize the acidic materials (H_2SO_4 and HNO_3) in the aerosol. By measuring both gaseous NH_3 and NH_4^+ contained in the aerosol, it should be possible to account for all of the NH_3. A simple model based entirely on meteorological factors should account for changes in total $NH_3 + NH_4^+$.

To the best of our knowledge, there have been no extensive NH_3 monitoring programs. There are several analytical methods for NH_3. The classical wet chemical method involves absorption in dilute H_2SO_4 followed by analysis with Nessler's reagent. However, large volumes of air must be pulled through the dilute H_2SO_4 solution and some methods require distillation of the NH_3 from the neutralized absorption solution. An alternative instrumental method has been reported by Hodgeson et al. (1971). The NH_3 is catalytically oxidized to NO in a heated tube and detected by a chemiluminescence NO detector.

Beginning in July 1973 the research staff should select and test one of the possible NH_3 wet chemical analytical methods. A preliminary program should start as soon as facilities at the five research sites are installed. At the same time, NH_4^+ should be measured in the hi-vol samples collected at the five research sites. This brief program should delineate the general NH_3 concentration changes as an air parcel passes through St. Louis. Examination of the direction and magnitude of the changes should enable the research team to determine the course the NH_3 monitoring program should take.

Final recommendations for the remainder of the RAPS should be completed by January 1974. The NH_3 monitoring network should be operational by July 1974. Investigation of the factors which determine the rate of absorption of NH_3 by aerosols will be much more difficult. Important variables will include relative humidity (RH), SO_2, NO_x, $SO_4^=$, and NO_3^- in the aerosol, which are already required for other research programs during the RAPS. However, since the absorption of NH_3 will be a necessary step for formation of aerosols containing both NO_3^- and $SO_4^=$, we do not recommend an extensive NH_3 monitoring and modeling program at this time. However, the recommendations of the research team could include such a program.

The CO Research Program

There are two possible CO sinks in the biosphere. Inman et al. (1971) have found that fungi in the soil rapidly absorb low concentrations of CO. It is also well known that CO is oxidized to CO_2 by OH radicals. Using estimated average values for the rate of CO uptake and OH concentrations, we estimated that the rates could be comparable. While better estimates of the OH concentration in urban atmospheres would be desirable, it is virtually impossible to obtain such data with present techniques. However, improvements could be made to the CO absorption rate measurements by using more sensitive CO detection techniques. Such measurements may be available by 1974. The research team should evaluate these data as part of the CO monitoring program.

Current transport models simulate fairly well the dispersal of CO from urban sources. However, verification has been by measuring CO concentrations downwind of the sources. Similar data will be collected throughout the RAPS by the Class A and B stations. As a further verification of the CO models, the mass flux of CO passing through a vertical plane downwind from the urban area should be measured. Ground level measurements can be made at the downwind Class A stations. Aircraft transects will be necessary to obtain vertical CO profiles. From these data it should be possible, by including meteorological data, to calculate the mass flux through the vertical plane. The simulation model can also calculate a value for the mass flux, based on input data, such as traffic patterns, stationary CO sources, and meteorological conditions. The model can then be validated by comparing the measured value with that calculated using the model. This experiment would be invaluable in evaluating the importance of CO sinks. The measurements should be made three times in 1974, once each in spring, summer, and fall. Three days of measurements each time should provide sufficient validation data.

At the same time, atmospheric samples should be taken at the research sites for later submission for C^{12}/C^{13} isotope ratio measurements. These data provide supplementary information as to the origin of atmospheric CO.

REFERENCES

Altshuller, A. P. and J. J. Bufalini, Photochem. Photobiol. 4, 97 (1965).

Altshuller, A. P. and J. J. Bufalini, Environ. Sci. and Tech. 5, 39 (1971).

Altshuller, A. P., D. L. Miller, and S. F. Sleva, Anal. Chem. 33, 621 (1961).

Bates, D. R. and P. B. Hays, Planetary Space Sci. 15, 189 (1967).

Bufalini, M., Environ. Sci. Tech. 5, 685 (1971).

Cadle, R. D. and M. Ledford, Air and Water Poll. Int. J. 10, 25 (1966).

Chamberlain, A. C., Int. J. Air Poll. 3, 63 (1960).

Cheng, R. T., J. O. Frohliger, and M. Corn, J. Air Pollut. Control Assoc. 21, 138 (1971).

Dunham, S. B., Nature 188, 51 (1960).

Endow, N., G. J. Doyle, and J. L. Jones, Air Poll. Control Assoc. J. 13, 141 (1963).

Esmen, N. A. and M. Corn, Atmos. Environ. 5, 571 (1971).

Ford, H. W., G. J. Doyle, and N. Endow, ibid. 26, 1336, 1337 (1957).

Gartrell, F. E., F. W. Thomas, and S. B. Carpenter, Am. Ind. Hyg. Assoc. J. 24, 113 (1963).

Gerhard, E. R. and H. F. Johnstone, Ind. Eng. Chem. 47, 972 (1955).

Germogenova, O. A., J. P. Friend, and A. M. Sacco, Bolt Beranek and Newman, Inc., Cambridge, Mass., Final Report, Contract Nos. CAPA-6-68(1-68) and CPA 22-69-29 for Coordinating Research Council, New York, March 31, 1970. PB 192102.

Glasson, W. A. and C. S. Tuesday, Environ. Sci. Tech. 4, 916 (1970).

Harkins, J. and S. W. Nicksic, presented at American Chemical Society Meeting, Division of Petroleum Chemistry Detroit, Mich., April 4-9, 1965 (reviewed in Bufalini, 1971).

Hill, A. C., J. Air Poll. Control Assoc. 21, 6 (1971).

Hodgeson, J. et al., "Application of a Chemiluminescent Detector for the Measurement of Total Oxided of Nitrogen and Basic Nitrogen in the Atmosphere," presented at the Joint Conference on Sensing of Environmental Pollutants, Palo Alto, Calif., 1971.

Inman, R. E., R. B. Ingersoll, and E. A. Levy, Science 172, 1229 (1971).

Inman, R. E. and R. B. Ingersoll, ibid. 21, 646 (1971).

Jaffe, L. S., Air Pollut. Control Assoc. J. 18, 534 (1968).

Johnson, W. B., F. L. Ludwig, W. F. Dabberdt, and R. J. Allen, "Development and Initial Evaluation of an Urban Diffusion Model for Carbon Monoxide," Paper No. 25 B, presented at the 64th Annual Meeting, American Institute of Chemical Engineers, San Francisco, California, November 28-December 2, 1971.

Johnston, H. S. and H. J. Crosby, J. Chem. Phys. 22, 689 (1954).

Johnstone, H. F. and D. R. Coughanowr, Ind. Eng. Chem. 50, 1169 (1958).

Junge, C. E. and T. Ryan, Quart. J. Roy. Meteorol. Soc. 84, 46 (1958).

Junge, C. E., Air Chemistry and Radioactivity, Academic Press, New York, New York, 1963, 123.

Katz, M. and G. A. Ledingham, National Research Council of Canada, in Effect of Sulfur Dioxide on Vegetation, NCR No. 815, Ottawa, 1939.

Klosterman, D. L. and J. E. Sigsley, Environ. Sci. Tech. 1, 309 (1967).

Kummler, R. H., M. H. Bortner, and L. S. Jaffe, Environmental Sci. Tech. 5, 1140 (1971).

Leighton, P. A., Photochemistry of Air Pollution, Academic Press, New York, 1961.

Levy II, H., Science 173, 141 (1971).

McConnell, J. C., M. B. McElroy, and S. C. Wofsy, Science 1 (1971).

McKay, H.A.C., Atmos. Environ. 5, 7 (1971) and references therein.

Moffat, A. J., J. R. Robbins, and A. R. Barringer, Atmos. Environ. 5, 511 (1971) and references therein.

Prager, M. J., E. R. Stephens, and W. E. Scott, Ind. Eng. Chem. 52, 521 (1960).

Renzetti, N. A. and G. J. Doyle, Int. J. Air Pollut. 2, 327 (1960).

Ripperton, L. A. and D. Lillian, Paper 70-122 presented at 63rd Annual Meeting, Air Pollution Control Association, June 14-19, 1970, Accession No. N71-11216.

Robbins, R. C., "Developments of methods for selective removal of oxidants from smog," Final Report, Project S-3200, Stanford Research Institute, for Agricultural Air Research Association, University of California at Riverside, January 17, 1961.

Robbins, R. C., R. D. Cadle, and D. L. Eckhardt, J. Meteor. 16, 53 (1959).

Robinson, E. and R. C. Robbins, "Sources, Abundance, and Fate of Gaseous Atmospheric Pollutants," Final Report SRI Project PR-6755 for American Petroleum Institute, Washington, D.C., February 1968.

Schvetzle, D., private communication, 1971.

Seinfeld, J. H., P. M. Roth, and T. A. Hecht, "A Kinetic Mechanism for Atmospheric Photochemical Reactions," Appendix B of Report 71SAI-9, Systems Applications, Inc., Beverly Hills, Calif., for EPA Contract NO CPA 70-148, May 1971.

Sethi, D. S., J. Air Pollut. Control Assoc. 21, 418 (1971).

Smith, B. M., J. Wagman, and B. R. Fish, ibid. 3, 5581 (1969).

Stevens, C. M., L. Krout, and D. Walling, "Carbon Monoxide of the Atmosphere for the Northern Hemisphere and from Natural Sources of the Biosphere," Annual Report to Coordinating Research Council, Contract CAPA 4-68(1-69) and U.S. Atomic Energy Commission, May 1, 1971.

Stevens, E. R., in *Advances in Environmental Sciences*, Vol. 1, ed. J. N. Pitts, Jr. and R. L. Metcalf, Wiley-Interscience, 1969, p. 119.

Tingey, D. T., "Foliar absorption of NO_2," M.S. Thesis, Dept. of Botany, Univ. of Utah, Salt Lake City, Utah, June 1968.

Urone, P., H. Lutsep, C. M. Noyes, and J. F. Parcher, Environ. Sci. Tech. 2, 611 (1968).

U.S. Department of Health, Education, and Welfare, Public Health Service, National Air Pollution Control Administration, Washington, D.C., "Air Quality Criteria for Carbon Monoxide," March 1970b.

U.S. Department of Health, Education, and Welfare, Public Health Service, National Air Pollution Control Administration, "Air Quality Criteria for Hydrocarbons," Publication No. AP-64, March 1970a.

U.S. Department of Health, Education, and Welfare, Public Health Service, National Air Pollution Control Administration, "Air Quality Criteria for Nitrogen Oxides," Publication No. AP-84, January 1971a.

U.S. Department of Health, Education, and Welfare, Public Health Service, National Air Pollution Control Administration, "Air Quality Criteria for Particulate Matter," Publication No. AP-49, January 1969.

U.S. Department of Health, Education, and Welfare, Public Health Service, National Air Pollution Control Administration, Washington, D.C., "Air Quality Criteria for Sulfur Oxides," January 1969b.

Wayne, L. et al., J. Air Pollut. Control Assoc. 21, 334 (1971).

Went, F. W., Proc. Nat. Acad. Sci. 46, 212 (1960); Tellus, 18(203), 549 (1966).

Westberg, K., N. Cohen, and K. W. Wilson, Science 171, 1013 (1971).

Wilson, W. E. and A. Levy, "A Study of Sulfur Dioxide in Photochemical Smog," Battelle Memorial Institute Report to the American Petroleum Institute, August 1968.

Chapter V

RESEARCH PLAN--EMISSION ESTIMATES

Requirements of the Emission Inventory System

An emissions inventory system is an essential feature of any comprehensive air pollution abatement program for an area containing a substantial number of sources. At the very least, it serves to establish the dimensions of the problem. It furnishes the basis for planning any emissions reduction program, whether or not linked specifically or quantitatively to air quality considerations, and thus is a necessary component of an air quality implementation plan. Finally, an emissions inventory must be employed in any attempt to predict air quality through use of atmospheric diffusion models, simple or complex. It is this last application that imposes the most stringent demands upon the emissions inventory, since the time intervals of interest are generally relatively short and the problem of producing an accurate inventory is thereby accentuated. A number of emission inventories have been carried out for various portions of the Greater St. Louis area, but they have been based on long-term average emission rates.

The emission inventory system must satisfy two basic requirements:

- The system must be applicable to the inventory of all sources and all pollutants, including any not currently being considered for immediate study or application of controls.
- The system must be adaptable to the needs of atmospheric diffusion modeling.

To satisfy the latter requirement, the inventory must be able to predict with reasonable precision the emissions of any given pollutant within any specified one-hour period. The inventory must therefore be given in terms of baseline values of pollutant emissions together with appropriate factors for adjusting the baseline emissions to the conditions of the specified time interval. In this form, it is probably better described as an "emission model." Determination of the baseline emissions will necessarily require an extended period of time, but the inventory system must permit ready correction of the baseline values for long-term changes. The adjustment factors for short-term temporal variations should require use of the least possible number of independent measurements to describe the conditions of the specified time interval.

Classification of Emission Sources

The broad classification of emission sources is presented in Table V-1. This scheme is intended to accommodate all possible sources and pollutants in a format that is structured according to the methodology that must be used to gather the data and according to the way that the information must be applied in diffusion modeling.

The primary division of sources into categories separates the stationary from the mobile sources, since these present radically different problems with respect to both emission inventories and modeling. In the secondary division, stationary sources are divided into area sources and point sources, whereas mobile sources are composed of area and line sources. Dividing the mobile sources into area or line sources is a matter of expediency. Well-defined and heavily traveled traffic arteries, such as freeways, can be treated as individual line sources. The more diffuse traffic on city streets can best be handled on an area basis.

The division of stationary sources into point and area sources is necessarily arbitrary. The point sources, or source units, are those large enough to warrant individual consideration. Area source units are, by contrast, ones having relatively small emissions, and they cannot for practical purposes be treated individually. The emissions from those small units existing in a given area are therefore aggregated and estimated from some factor, such as the consumption of fuel within the specified area. In diffusion modeling, the aggregate emission may either be considered as discharged from the center of the pertinent area, or as distributed uniformly over it, depending on the requirements of the model.

The criterion of size for the definition of point source units is relative, and is related mainly to the precision desired for the inventory and for the diffusion estimates derived from it. A unit emitting a small absolute quantity of pollutant material may in fact be an important point source if it nevertheless contributes an appreciable fraction of the total emission of that specific pollutant into the region. A given source unit may be relatively insignificant with respect to one pollutant of interest, and at the same time be a very large emitter of another pollutant.

Table V-1

CLASSIFICATION OF SOURCES FOR EMISSION INVENTORY

Source Category	Stationary Sources				Mobile Sources	
Source Subcategory	Area Sources		Point Sources		Area and Line Sources	
Source Process	Combustion	Noncombustion	Combustion	Noncombustion	Combustion	Noncombustion
Source Units	Commercial Institutional Residential Small Industrial Fuel Use Space heaters Water heaters Boilers Waste Disposal Incinerators	Commercial Small Industrial Venting of organic vapors (dry cleaning, painting, gasoline storage and handling, food preparation	Utilities Power plants Municipal incinerators Industrial Boiler and power plants Indirect-fired air and process heaters Stationary internal combustion engines Stationary gas turbine engines Incinerators	Industrial Direct-fired process units All other industrial processes, material storage and handling	Surface Vehicles Passenger cars Trucks and buses Commercial vehicles Railroads Vessels Off-highway vehicles and equipment Aircraft Piston engines Gas turbines	Surface Vehicles and Aircraft Venting of fuel vapors Wear of tires and brakes
Pollutants	Gases and Vapors SO_x NO_x CO Hydrocarbons and derivatives HCl HF Odors Particulates Fly ash and its specific chemical components Smoke	Gases and Vapors Organic vapors (solvents, gasoline) Odors Particulates Organic aerosols Smoke	Gases and Vapors SO_x NO_x CO Hydrocarbons and derivatives HCl HF Odors Particulates Fly ash and its specific chemical components Smoke	All pollutants	Gases and Vapors SO_x NO_x CO Hydrocarbons and derivatives Odors Particulates Smoke Lead Oil aerosols Derivatives of fuel additives	Hydrocarbon Vapors Particulates Organic Inorganic

The decision as to whether a given emitter should be considered a point source is also dependent on the location of the receptor. (The receptor may in this case be considered as the point of measurement of the pollutant atmospheric concentration.) If the receptor is located nearby, even a relatively small emitter may have to be considered as a point source. On the other hand, if the receptor is located at a relatively great distance from the same emitter, the individual influence of the latter on the pollutant concentration at the point of measurement may be indistinguishable from the influence of a mass of other small sources.

The potential accuracy of an emission inventory or model, and hence of diffusion models employing its output, is largely related to the fineness of resolution of the inventory; that is, to the degree to which individual source units are specified. The actual accuracy required will depend on the purposes for which the inventory is used and the particular circumstances under which it is applied. In general, the degree of source resolution actually employed in diffusion modeling will probably be less than that attained in the emission inventory. However, this cannot be assumed to be true in all cases, and it will be necessary to be reasonably conservative in specifying source resolution in the inventories. Workers engaged in diffusion modeling should undertake to indicate the maximum resolution that they will require, although compromises between the accuracy and the data-gathering effort will often be required. Diffusion modelers will also have to exercise judgment in deciding what part of the source resolution available to them should actually be employed in any given study or set of circumstances.

Agricultural activities give rise to a number of atmospheric pollutants, which generally come from emission sources in the categories defined in Table V-1, or in directly analogous categories. For example, engine exhausts from tractors and other powered agricultural equipment fall directly into the category of mobile (or semimobile) combustion sources. Dusts raised by operation of agricultural equipment are comparable with those produced by materials handling and storage in industrial operations. Odors from cattle feed lots are likewise comparable with those from various industrial processes. The wind-blown dust from farm land is analogous to that from material storage piles at industrial plants, and its amount is closely related to farming practice. Burning of agricultural wastes parallels waste incineration in urban areas.

On the other hand, the application of agricultural chemicals such as insecticides, fungicides, and herbicides, is in some degree a special case. The agents are specific in their chemistry and effects. They are also deliberately disseminated into the atmosphere, and become air pollutants to the extent that they drift beyond the area of intended application.

Source Processes

The source processes are conveniently classified as either combustion or noncombustion processes. Combustion processes are defined as those in which the pollutants are produced exclusively by the burning of fuels or of solid or liquid wastes. They include all those processes in which there is indirect transfer of the heat produced (e.g., boilers, indirect-fired air heaters) as well as incinerators, internal combustion engines, and gas turbines. Noncombustion processes comprise all other pollutant sources not falling under the specific definition of combustion processes. They include operations in which combustion takes place, but in which part or all of the pollutants emitted arise from operations other than the burning of fuel or wastes. Examples are those processes in which the products of fuel combustion come into direct contact with materials being processed such as calcining of materials in kilns.

By far the largest number of pollutant sources, stationary and mobile, are combustion sources. In particular, combustion processes comprise the most important area source units, and estimates of emissions from these numerous contributors can be made from estimates of fuel consumption. The most important noncombustion sources are industrial, and are generally point sources.

Source Units

Stationary Sources

Area Sources--The stationary area sources are composed of residences, commercial establishments and activities, institutional establishments (e.g., schools, hospitals, and public buildings), and some small industrial (manufacturing) establishments. Most of the pollutants arise from the combustion of fuels used for space and water heating and steam generation, and from combustion of wastes in small incinerators. However, the commercial category of sources includes some noncombustion sources--primarily the venting of organic vapors from gasoline storage and handling by distributors and service stations, dry cleaning, painting, degreasing, and application of asphalt to roofing and pavement. Odors and smoke from large-scale food preparation, such as commercial barbecue pits, sometimes constitute local nuisances, but are unlikely to contribute significantly to total area emissions.

Industrial establishments in this category are composed mostly of small manufacturing operations that employ fuels primarily for space heating, or that burn wastes.

Occasionally, individual source units in the residential, commercial, or institutional classes may be large enough to constitute point sources. Possible examples are the central heating plants of a large apartment complex or of a university.

Point Sources--The stationary point sources comprise an extremely wide variety of public utility and industrial (manufacturing) activities. They may also include a few commercial and institutional sources, as noted above.

When industrial plants are considered, it may be desirable to define "sources" as entire plants rather than process units, at least from the standpoint of data gathering. Some operations within an industrial plant, such as space heating, might individually be of a scale to rank only as contributors to area sources. Nevertheless, the data should be accumulated initially as for other point sources, however it may be treated thereafter.

Utility power plants include all forms of thermal power generation systems: gas-, oil-, and coal-fired boilers, gas turbines, and diesel engines. They may also include some noncombustion sources, such as coal grinders and driers, and coal- and ash-handling systems. Gas turbines and internal combustion engines are commonly employed as prime movers for compressor stations on natural gas transmission lines. Municipal incinerators and most industrial incinerators for solid and liquid wastes will also fall in the category of point combustion sources.

Industrial point combustion sources will include steam boilers for generation of either electrical power or process steam, indirect-fired air and process heaters, and stationary internal combustion and gas turbine engines. Industrial noncombustion processes will best be treated exclusively as point sources. Even in cases where their emissions prove to be small, this condition will not generally be obvious without examination of the particular instances. The industrial noncombustion source category can be used to encompass virtually all stationary pollutant sources not specifically defined elsewhere, including sources as diverse as cement kilns, windblown dust from material storage piles, and vents of organic chemical manufacturing processes.

Mobile Sources

Mobile sources cannot, by their nature, be considered in a practical way as individual units. However, they must be broken down by classes to permit application of appropriate emission factors. The classes of surface vehicles and aircraft are presented in Table I. The class of off-highway vehicles and equipment includes tractors, earth-moving equipment, and so on.

With respect to emissions inventory procedure, the distinction between combustion and noncombustion sources is of far less importance in the case of mobile sources than in that of stationary ones. Whereas the stationary source units in the two categories are usually physically as well as conceptually distinct from one another, the mobile source units fall in both categories. The same inventory procedures and operations that may be used to estimate combustion-related pollutants from vehicles can also be used simultaneously to provide data on emissions of the noncombustion pollutants, since volatilization of fuel and generation of particulates by wear of brakes and tires can, like the emission of engine exhaust, be related to the mileage that a vehicle is driven.

Pollutants

The pollutants from noncombustion sources may include anything (see Table V-1). The major pollutants from combustion sources are relatively few in number, although many other materials may be present in relatively small quantities. Broadly, there are two groups of pollutants from combustion of fuels. First, there are those arising from the oxidation of the atmospheric nitrogen itself (nitrogen oxides) and from incomplete combustion of the hydrocarbon or carbon fuels, such as unburned hydrocarbons, aldehydes, carbon monoxide, and soot. Second, there are the pollutants arising from the materials in the fuel other than hydrogen and carbon--sulfur, chlorine, selenium, mercury, iron, alumina, silica, and so on. These latter materials may either be chemically combined with the hydrocarbon materials, or merely be mixed with the fuel (e.g., the ash in coal).

The incineration of solid or liquid wastes can, of course, be responsible for the emission of a very wide variety of pollutants, depending on the composition of both the combustible and noncombustible portions of the waste.

Mobile sources primarily emit pollutants associated with combustion of fuels, including fuel additives such as tetraethyl lead. However, they are also responsible for emission of noncombustion pollutants. Hydrocarbon vapors are emitted by the venting of fuel tanks and by the dumping of fuel from aircraft. Motor vehicles generate dust by wear of tires and brakes.

Factors Affecting Emission Levels

Stationary Sources

Area Combustion Sources--The greater portion of the fuel consumed by area combustion sources is used in space and water heating and in cooking. The rate of fuel consumption therefore exhibits pronounced diurnal and seasonal variations as well as variations related to ambient temperatures. Fuel consumption peaks appear over periods of one to three hours. Fuel consumption patterns at commercial, institutional, and small industrial operations depend on whether operations are carried on only during the day, or during nights and weekends as well. They therefore vary from the consumption patterns of residential resources. Both the nature and the amounts of pollutants generated vary with the type of fuel used, which is in turn undergoing changes throughout many cities, including St. Louis. Individual residences and small apartments increasingly tend to use natural gas, whereas large apartment houses and commercial, institutional, and industrial operations are relatively more likely to use oil or coal. Emissions are also related to the size and type of combustion equipment, with the effect [illegible] factors on emissions probably being most pronounced in coal-[illegible]

Practices employed in waste incineration [illegible] ly, where incineration is used at all.

Area Noncombustion Sources--The most important of the area noncombustion sources are those emitting organic vapors. The emissions of gasoline vapors from automobile service stations will vary with the day of the week and the hour of the day, as the volume of business fluctuates. The emission of cleaning fluid vapors from dry cleaning establishments should be relatively more constant during actual working hours. Solvent vapor emissions from the painting of products in small manufacturing establishments will have a similar temporal pattern. On the other hand, the solvent vapor emissions from the painting of buildings will be irregular with respect to both the time and the location of the operation.

Point Combustion Sources--Utility power plants handle a fluctuating generating load that usually follows a fairly well defined cycle.[1] The typical load passes through a minimum in the early morning hours and reaches its peak value in the afternoon. The absolute levels are functions of the season of the year, as are the hours at which the minimum and maximum values are reached. The daily peak load varies with the average daily temperature,[1] and presumably could be even more closely correlated with the hourly mean temperature. The daily peak load increases with lowering of the average daily temperature during the fall, winter, and spring, evidently reflecting increased space heating loads. It increases sharply with increasing average daily temperature during the summer, and more moderately in the fall, showing clearly the influence of the air conditioning load.

The pattern of emissions from power plants will reflect the generating load cycle, but the absolute emission levels at any given plant will be set largely by the fuel and the air pollution equipment used. Some power plants, at least, may be able to shift from one fuel to another during various periods, and a general trend toward use of low-sulfur fuels is probable during the early phases of control implementation plans. The performance of fly-ash control equipment will be the most important factor determining emissions of particulate matter from coal-burning plants.

Industrial power and steam plants will usually display load patterns substantially different from those of the public utility plants serving the general area. Where the manufacturing plants are operated on a 24-hour basis, both the electrical power and the steam required for process use should in general represent fairly steady loads. The power and steam required for air conditioning and space heating will, of course, superimpose a fluctuating load on that resulting from process requirements.

Depending on the type of manufacturing, the process loads may exhibit cyclic behavior, perhaps over short intervals (electrical power loads in a steel rolling mill). However, the hourly average demand may not vary much from hour to hour.

Point Noncombustion Sources--Point noncombustion sources comprise such a diverse collection of manufacturing operations that little of a general nature can be said of the factors affecting their emissions. Most of the large emitters operate fairly steadily on a 24-hour basis, but there are a number of batch processes whose emissions show a cyclic behavior over short intervals even though their longer term averages are relatively constant. An example of the latter category is the pushing and quenching of coke at coke oven plants.

---

[1] J. J. Roberts et al., A Multiple-Source Urban Atmospheric Dispersion Model, Argonne National Laboratory Report No. ANL/ES-CC-007 (May 1970).

When the point noncumbustion sources are inventoried, an essential part of the task will be determination of the pattern of operation so that short term emissions can be estimated.

Mobile Sources

The average emissions from automobiles, expressed as mass emitted per mile of travel, are functions of the average vehicle speed. The emissions of hydrocarbons and carbon monoxide per mile driven tend to decrease as driving speed increases until about 70 mph is reached, then increase again. At the lower driving speeds, the approach to the idling mode results in a richer engine mixture, with accompanying higher emissions. Also, in actual highway travel, acceleration and deceleration tend to be more prevalent at the lower than at the higher speeds, and lead to relatively higher emissions. At very high speeds, also, the engine mixture tends to be richer.

In contrast to the emissions of carbon monoxide and hydrocarbons, the emission of nitrogen oxides can be expected to increase generally with increase in driving speed.

The volume of traffic of course exhibits a very marked diurnal pattern, with the patterns for Saturday and Sunday differing from those for weekdays.[2] The pattern of use of various streets and other traffic arteries also varies at different hours of the day. There is also some seasonal variation in traffic volume. Since average vehicular speeds on the different types of traffic arteries vary, so do the emission rates.

Obviously, the installation of emission control devices will in the future be a major factor affecting emissions from motor vehicles. Estimation of emissions must include allowances for the distribution of ages among the automobile population.

---

2. Ludwig, F. L., A. E. Moon, W. B. Johnson, and R. L Mancuso, A Practical, Multipurpose Diffusion Model for Carbon Monoxide, Stanford Research Institute Final Report to CRC/NAPCA, Contract No. CAPA-3-68/CPA 22-69-64 (September 1970).

Inventory Procedures and Accuracy

Accuracy of Estimates

Atmospheric diffusion models are based on the assumption that the predicted atmospheric concentration of a pollutant is directly proportional to the source strength (i.e., the estimated pollutant emitted per unit time from the source). Therefore, for the case of a single emission source, the error in the predicted concentration will also be directly proportional to any error in the estimate of the emission. If there are multiple pollutant sources, the error in the predicted concentration will still be directly proportional to any systematic error in the total estimated emission. However, random errors in the estimates of the component emissions will tend to cancel each other, reducing their effect on the predicted concentration. Obviously, the degree to which the random errors actually do cancel depends on the weighting of the individual component estimates (and their errors) in the total.

In the limit, of course, the accuracy of predictions made from diffusion modeling cannot exceed the accuracy of the input data on pollutant emissions, and may be less because of other and possibly larger errors in other input data. In the testing of diffusion models, it is essential that the accuracy of the emission estimate be such that it will not be the ultimate limiting factor in the accuracy of the tests. An essential part of the research program will be an examination of the magnitude of the errors introduced into the emission inventory by the procedures and assumptions used, and of the effects of these errors on the validation of the diffusion models. Another essential research task will be to devise inventory procedures that will minimize these errors without adding unreasonably to the time and costs required for execution of the inventory.

Long term emission inventories (e.g., those yielding annual average emission rates) can be made with a relatively high degree of accuracy, limited primarily by the level of effort that can be devoted to data gathering. However, several man-years of effort will generally be required to obtain the data for a large metropolitan region, such as St. Louis. Furthermore, when emissions are to be estimated for some specific area and for a short interval of time, primary data leading directly to emission estimates may simply not exist; indirect means of estimation must be used. The other inputs to the diffusion model, primarily meteorological data, will be obtained on a real-time basis, as will be the measured atmospheric pollutant concentrations used to check the predicted values. However, essentially no primary emission data can be gathered on a real-time basis. A stated goal for the modeling program is to treat time intervals as short as one hour. For intervals of one hour, or even of several days, it is possible only to measure certain variables that may be used to adjust baseline values of emission rates to the conditions of the particular period of concern. In the case of fuel combustion for space heating, the pertinent variables may include the atmospheric temperature, the hour of the day, and the season of the year, which will all affect the consumption of fuel. In a power plant, the consumption of fuel can be related to the amount of electrical power generated. Once the amount of fuel consumed is known, the estimate of the emission still depends on another predetermined or assumed quantity, the emission factor. Emission factors can represent quite accurately the magnitudes of long-term, average emissions. On the other hand, the use of average emission factors can easily result in an error of twofold or more when a short time interval is involved. This is particularly true of noncombustion processes. Additional, subsidiary factors descriptive of the emitting process may therefore have to be applied to the emission factor for an important point source.

In the Chicago Air Pollution Systems Analysis Program conducted by Argonne National Laboratory,[1] algorithms were developed to simulate the hourly variation of utility power plant electricity generation and pollutant output. Although ANL used actual hourly megawatt production as input data in its validation tests of the diffusion model, the algorithms are used in routine applications of the model. ANL also developed algorithms to simulate the emissions from other point combustion sources.

Although similar algorithms can be derived for point sources in the St. Louis area, it appears advisable to employ specific and directly obtainable data to the maximum degree possible during model validation tests. Each simulation necessarily involves certain potential errors that should, if possible, be avoided in the validation tests even though they may be acceptable in more routine applications of diffusion models.

It will also be necessary to develop more accurate emission factors for large individual emission sources, probably including values specific to some of the St. Louis operations. For small, numerous units, such as gas-fired space heaters, average factors can be developed from tests of typical units, either in the St. Louis area or elsewhere. However, for at least part of the large sources, such as power plant boilers, it will probably be necessary to develop data from sampling of the specific units in the St. Louis area. Accurate information must also be obtained on the efficiencies of air pollution control systems in use.

Procedures

Execution of Emission Inventories--The emission inventory for the St. Louis region, which contains a complex combination of many different sources, will actually consist of a number of subsidiary inventories that can, and in some cases probably should, be conducted separately, since some of them call for different kinds of talents and experience on the part of the personnel carrying out the work. In the case of point sources, it will be necessary to make a source-by-source survey, employing direct interviews. Mailed questionnaires cannot be expected to yield the detailed information needed. Because of the extreme diversity of the industrial noncombustion sources, in particular, it is not practical even to produce an adequate, generalized questionnaire. Individuals conducting the interviews must make themselves reasonably familiar with the process technology of the pollution sources.

It is essential that whatever agency conducts the emission inventories have full legal authority to require submission of complete information from all pollutant emitters. Otherwise, the required information simply will not be obtained, as has been demonstrated by the experience of agencies that have sought to obtain information on a voluntary basis. However, it is obvious that the required data may include industrial information that either is, or may be claimed to be, confidential or proprietary. It is here assumed that the EPA will have authority under the Clean Air Act of 1970 to collect the information needed, but it is not clear whether the task can be delegated to a contractor or even to state or local government agencies. For the present it is assumed that collection of the information will have to be carried out by EPA personnel. Furthermore, since analysis of the data and their incorporation into emission models will also require contact with the same confidential information, it must also be assumed that only EPA personnel will be able to carry out that work.

In any case, it is essential that a single agency carry out (or, at least, prescribe and supervise) the collection of inventory information for the entire air quality region under study, without regard to state or local governmental boundaries. Only in this way can uniform procedure and treatment of information be assured.

Point Sources--Previous emission inventories of the St. Louis area have identified about 100 to 150 point sources of one or more of the following major pollutants: sulfur oxides, particulates, carbon monoxide, hydrocarbons, and nitrogen oxides. A more refined inventory and inventory procedure necessary to support the diffusion modeling program may require a finer solution of sources, with specification of a much greater number of point sources. In particular, the number of point sources treated must be increased once consideration is given to the noncombustion sources of particular pollutants not specifically indicated in the listing given above.

From data of the U.S. Census of Manufacturers and other sources, it appears that there may be within the St. Louis Interstate AQCR as many as 1000 or more manufacturing establishments, utility plants, and other operations that might be considered point sources with respect to some pollutant or other. Some of these establishments may have a number of emission points differing with respect to pollutant emitted, stack height, and stack gas temperature and exit velocity, as well as with respect to physical location. A steel plant may easily have several dozen emission points, but not all of these will necessarily qualify for classification as point sources with respect to any given pollutant being considered. Actual or potential point sources must be identified at a relatively early date in the inventory program, but in a few cases information from the survey itself may be necessary to establish definitely whether the sources should actually be treated as point sources.

The data collected for the point sources must include not only the emissions, but also the information necessary to permit calculation of the diffusion of the pollutants emitted--stack height and diameter, stack gas exit velocity and temperature, and information for adjusting the latter two factors for variations in the source operating conditions.

For the individual point sources--or for groups of point sources, if possible--it will be necessary to establish a baseline level of emissions that can be adjusted to the circumstances of a particular hour. The baseline condition can be chosen for convenience, and need not be the same for all sources. However, the inventory system must provide for review at frequent intervals to maintain current information on major systematic changes in the sources, such as changes of fuel or installation or upgrading of control equipment.

The factors used to adjust the baseline inventory values to the conditions of the desired one-hour period should be few in number, and should as far as possible consist of (or be derived from) routinely recorded operating data. In the case of power plants, this will be the electrical energy generated. For industrial boilers, it may be the quantity of steam produced. Provisions must be made to collect such information rapidly and with a minimum of effort.

Because industrial sources are extremely diverse and information about them is relatively less accessible than for utilities, the derivation of adjustment factors for their emissions will be particularly dependent on the survey interviews. One of the tasks of the inventory will be to decide which industrial sources will have to be checked specifically with respect to their operations during the given hour of a model validation test, and which can safely be assumed to be operating in their steady, "normal" condition. Adjusting the baseline emission rates to the circumstances of a particular hour may be relatively easy for some processes that operate steadily on a 24-hour basis, but very difficult for certain other processes that operate irregularly, or on a batch or cyclic basis.

Stationary Area Sources--Area combustion source units cannot be surveyed individually, and their emissions can be estimated only through estimation of their fuel consumption and consumption patterns. Emission factors characteristic of the general type of combustion equipment and of the type of fuel must be applied to the quantity of fuel burned to obtain estimates of the amounts of pollutants emitted. The greatest problem lies in determining the rates of consumption of each kind of fuel in a given area during the specific, short (one-hour) time intervals of interest.

In the case of natural gas, the overall consumption pattern for large areas is well known by utility companies supplying the gas. However, the degree to which the consumption of gas can be broken down by areas within a city or other region is uncertain. Practice in locating metering stations is not uniform. Furthermore, the gross usage of gas in the area supplied from a given metering station will in general represent the overall pattern of consumption for a variety of emission sources, possibly including some point sources being accounted for separately. Where oil and coal are used for space and water heating in area sources, information on fuel consumption is necessarily limited to data on the total quantities consumed, generally over periods ranging from weeks to months.

Determination of the short-term patterns of consumption of the different types of fuels by area emission source units will call for relatively extensive field sampling surveys, such as have been made in Chicago by ANL and described by Roberts at al.[1] in the report cited above. Models must then be developed to represent the emissions from each type of source in terms of the amount and kind of fuel used, the time of year, the time of the day and day of the week, and the ambient temperature. Survey information must be used to derive the number of each type of source unit in a given area.

The emission models for the stationary area sources must be reviewed at frequent intervals to take into account changes in the types of fuel used, such as substitution of natural gas for oil or coal, or of oil for coal.

The average emissions (primarily organic vapors) from area noncombustion sources can be estimated by applying appropriate emission factors to the quantity of gasoline handled, the amount of cleaning fluids consumed, the amount of paint applied, and so on. However, it will be necessary to determine the actual temporal pattern of operations to permit estimation of the emissions during a given short time interval. To do this will require surveying the operations at a representative sample of establishments or activities of each type.

Mobile Line and Area Sources--A number of diffusion models have been developed for estimating the spread of pollutants from vehicular traffic, and Ludwig et al.[2] have already applied one to the St. Louis area. Corresponding emission inventories have been developed for use with these diffusion models, and an example is presented in the report by Ludwig et al.[2]

The automotive emissions can be divided into two components: (1) primary network link emissions from vehicles traveling on the network of major arterial streets and freeways, and (2) secondary background emissions from vehicles traveling over the less densely traveled local and feeder streets. The primary network links comprise the sections of major arterial streets and freeways between intersections with other major arterials and freeways. The emissions from each link, which constitute a line source, can be computed from the estimated traffic flow and emission factors characteristic of the vehicles and their average speed of travel. The vehicles traveling on streets not represented by the primary network are treated as contributors to area sources. The area under study is divided into squares according to the chosen grid system, and the emissions from the local street travel in a given square are assumed to emanate uniformly from that square.

The foregoing basic inventory system can be applied in the Regional Air Pollution Study. The necessary refinements in the inventory will deal with the methods for estimating the flow of traffic and the emissions from the vehicles. One of the more critical and difficult problems will probably be the determination of truly representative emission factors for vehicles operating under each set of circumstances.

The data on total volume of traffic passing over the primary network links can, for the most part, be obtained from the traffic departments of the city or county governments. Data on the diurnal variations in traffic flow can be obtained from the urban transportation studies made under terms of the Federal Aid Highway Act. However, obtaining an adequate definition of the diurnal variation in traffic will probably require that some special measurements be made. Allowance must also be made for the growth in the volume of traffic over the period of the Regional Study, and this will probably call for special measurements. The accuracy of forecast traffic data is uncertain.[2]

Special measurements and driver surveys will probably also be necessary to determine the patterns of secondary automotive traffic and its emissions.

The patterns of operation of public transportation facilities--including buses, trains, and aircraft--are easily determined. In some previous inventories, airports have been treated as point sources. However, for the purpose of short-term diffusion modeling, aircraft taking off and landing are probably better treated as intermittent line sources. The method of treatment of emissions from aircraft flying over the area at relatively high altitudes remains to be decided.

The emissions from off-highway vehicles and mobile equipment, such as tractors and earthmoving equipment, are probably a very minor part of the total and of little influence in the immediate area where the equipment is working.

Emission Model

The emission model will consist of algorithms developed from the emission inventories for all the sources and pollutants in the St. Louis area. Input to the algorithms of appropriate data descriptive of the particular time interval of interest will yield the corresponding emissions estimates. The model must supply not only the mass emission rate for the pollutant in question, but also the information on stack exit velocities and thermal emissions that are essential to estimating the effective plume rises employed in the diffusion models.

When established in this form, the emission model will be readily adaptable to a variety of purposes, the first in order of priority being, of course, the validation testing of diffusion models. A second important application will be in the testing of control strategies, although the validity of such studies will hinge on prior validation of the diffusion model employed.

Control Strategy Studies

Except for preliminary training exercises, meaningful control strategy studies using the diffusion and emission models must wait upon experimental demonstration of the validity of the diffusion model or models. The validity of the diffusion model must in this case be understood to mean the ability of the model to predict atmospheric pollutant concentrations within some specified degree of accuracy. The first diffusion models tested may not meet the requirements for accuracy necessary to make the results of control strategy studies significant, even though they may be useful for other purposes.

Unless the alternative control strategies being considered in a given case are widely different (e.g., overall emission reductions of 50% and 90%), an air quality model will have to be highly accurate to predict whether the more lenient of the alternatives will actually meet the specified air quality standard. For example, an error of ±25% in prediction of the actual atmospheric concentration may, in a simple case, permit one to decide whether the overall control efficiency should be 95% rather than 90%, which is a substantial difference in required performance. In more complex cases, and where proposed control efficiencies are closer together, prediction errors will have to be reduced below ±25%.

Inventory Schedule

In principle, it would obviously be desirable to inventory as many of the pollutant emissions as possible at the same time, to avoid repetitions of interviews and other duplications of effort. In practice, it will probably be necessary to limit the number of pollutants covered in any one inventory. The development of basic information essential to the inventory of certain pollutants may lag substantially behind the need to inventory other pollutants. For example, emission factors for nitrogen oxides are much less well established than those for sulfur oxides, and are far more difficult to determine experimentally or to predict. For numerous industrial noncombustion processes, emission factors for some pollutants will have to be determined experimentally before statistical data on production operations can be used with confidence to estimate emissions.

Since sulfur dioxide and carbon monoxide are the pollutants that will be used in the first tests of air quality models, the initial emission inventories will be focused upon them. The tests with sulfur dioxide essentially treat the case of gaseous pollutant diffusion from stationary sources, since motor vehicles are only relatively minor emitters of sulfur dioxide. On the other hand, motor vehicles contribute by far the largest portion of the total emission of carbon monoxide.

Tests of diffusion models for photochemically reactive materials (hydrocarbons and nitrogen oxides) will take longer to develop and execute than those for relatively nonreactive compounds, such as sulfur dioxide and carbon monoxide. Not only are the diffusion models themselves more complex, but developing the inventory information is more difficult. Emission factors for both hydrocarbons and nitrogen oxide are far less well established than those for sulfur dioxides, for example, and considerable experimental work will have to precede diffusion model tests.

As is discussed in Chapter III, the testing of diffusion models for particulate matter, poses special problems and is probably better handled in a different way. Modeling the diffusion of the particulate matter actually being emitted from various sources will therefore be carried on as a practical field demonstration of the technique rather than as a primary test of the model. Completion of the particulate inventory, including acquisition of the data on particle characteristics necessary for successful modeling, will be scheduled for a later date in the study.

Chapter VI

RESEARCH PLAN--ECONOMIC AND SOCIAL IMPACT STUDIES

Introduction

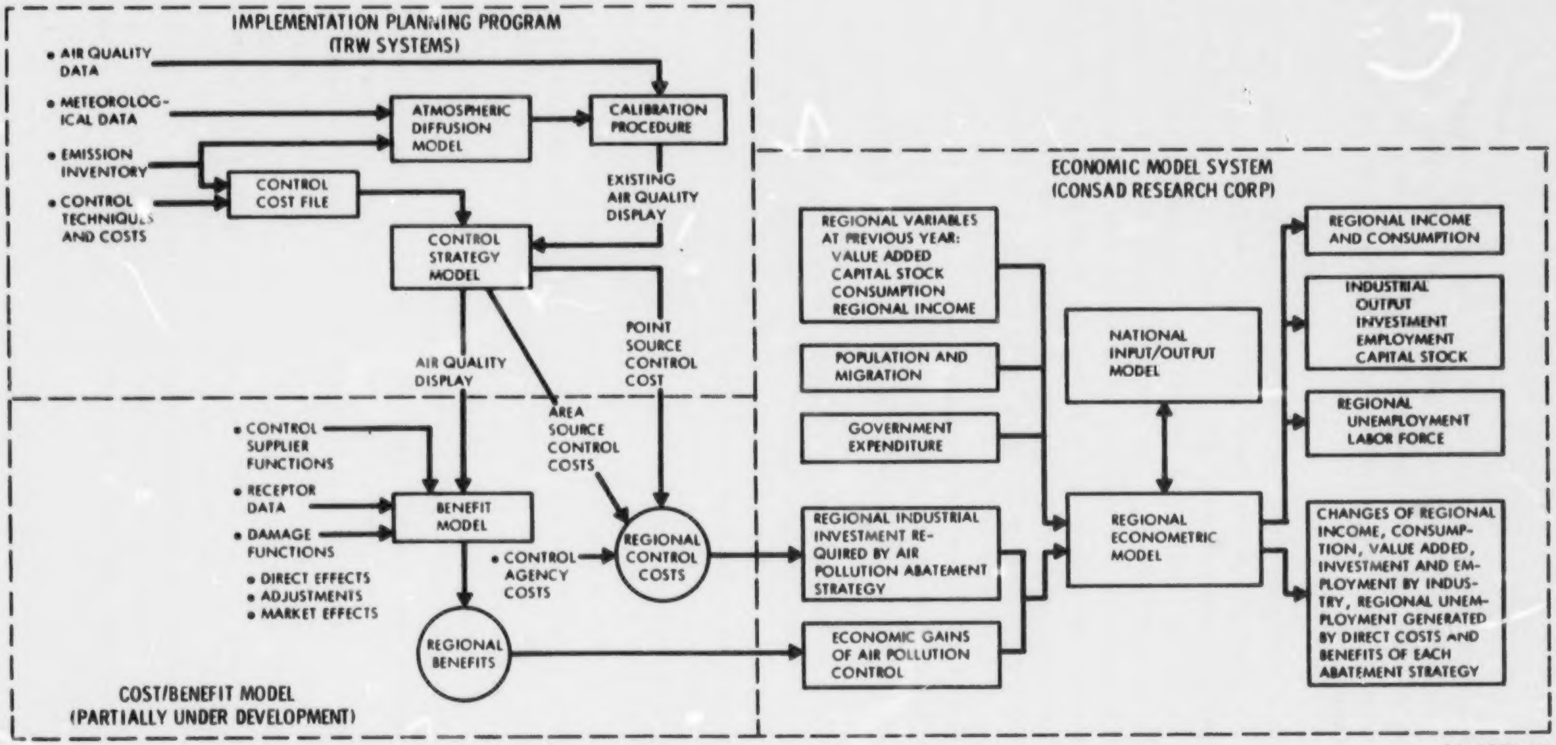
Air pollution problems are concerned not only with engineering, chemistry, physics, and meteorology, but also have large-scale impacts on the economic and social life of a community and a nation. The Regional Air Pollution Study provides an unparalleled opportunity to accumulate information on these community air pollution impacts because, if the necessary economic and social data are collected within the RAPS area, they can be related to detailed air pollution concentrations and exposure information. This correlation between detailed physical and social information on air pollution within the community should lead to an increased understanding and more accurate assessment of the actual impact of air pollution on the life of a community. Once a factual assessment of air pollution impact is available, air pollution control operations can be directed toward the most critical problem areas through new and revised control strategy programs. These programs can be judged not only on the basis of their impact on pollutant concentrations within the area but also upon their costs and expected benefits. Thus, this type of analysis can lead to control strategies that are judged upon their least cost maximum benefit performance as well as upon their performance relative to established air quality criteria.

There are several obvious costs of air pollution to the community. These include damage to health, damage to property through corrosion and staining, and reduced property values due to the unwillingness of buyers to accept obvious impacts of air pollution, such as increased dust fall, odors, and the like. Within a community the obvious benefits of air pollution control are reductions in the costs that are attributed to adverse pollutant levels. However, air pollution control has certain well-identified costs; these include the cost of operating a control agency, the cost to industry and to the public of buying and operating pollutant control equipment, and the increased cost of goods and services produced by industries where air pollution control significantly increases the operating overhead expenses of the operation. All of these factors--the damage due to air pollution, the benefits resulting from air pollution control, and the costs of air pollution control--are interwoven in a detailed fashion in the economic structure of any complex community. These factors are also related in a very detailed fashion to the meteorological and atmospheric pollutant patterns within the urban area. Figure VI-1 illustrates the three model components that might be considered necessary to define completely the impact of air pollution and its control on a community. As illustrated in this figure, the air quality and meteorological situations within an urban area impact on control strategies and in turn lead to community benefits and community costs. On a regional basis, both benefits and costs of air pollution control lead to industrial investments and economic gains for the community, which again are related to the total economic structure of the regional area.

The RAPS research program provides an ideal situation within which to collect specific costs and benefit data, such as source control costs, pollutant receptor distributions, and information on various air pollutant damage functions. The RAPS air sampling network and the RAPS research program will provide a vehicle within which the assessment of pollutant concentrations and pollutant exposures can be made for correlation with the observed or calculated costs and benefits. Although most of the needs of the economic cost and benefit program for detailed air quality and exposure data can probably be met by the regular air sampling program planned for the RAPS operation, it may be necessary in some cases to design special experimental or data-gathering projects to meet the needs of specific economic or pollutant damage programs.

Human and Social Factors

Within the area of human and social impact, factors dealing with the epidemiology and other health effects of air pollution must be considered to be of great importance. These research studies on health effects would involve not only the gathering of data on health factors and epidemiology for various population groups within the community but also the assessment of the exposure of these population groups to air pollution on the basis of models of pollutant distribution that are available or will be available from other aspects of the RAPS research program. After an initial assessment of health effects and their relationship to modeled air pollution exposures, it is likely that additional exposure information over particular population groups will be desirable. A network of transportable air pollutant monitoring stations can gather specific information in the neighborhood of the population groups under study. Through the use of adequately developed models and historical data on emissions within the RAPS experimental area, long-term exposures of subjects and population groups can be calculated with much greater


IMPLEMENTATION PLANNING PROGRAM (TRW SYSTEMS)
• AIR QUALITY DATA
• METEOROLOGICAL DATA
• EMISSION INVENTORY
• CONTROL TECHNIQUES AND COSTS
ATMOSPHERIC DIFFUSION MODEL
CALIBRATION PROCEDURE
CONTROL COST FILE
EXISTING AIR QUALITY DISPLAY
CONTROL STRATEGY MODEL
AIR QUALITY DISPLAY
AREA SOURCE CONTROL COSTS
POINT SOURCE CONTROL COST
• CONTROL SUPPLIER FUNCTIONS
• RECEPTOR DATA
• DAMAGE FUNCTIONS
• DIRECT EFFECTS
• ADJUSTMENTS
• MARKET EFFECTS
BENEFIT MODEL
• CONTROL AGENCY COSTS
REGIONAL CONTROL COSTS
REGIONAL BENEFITS
COST/BENEFIT MODEL (PARTIALLY UNDER DEVELOPMENT)
ECONOMIC MODEL SYSTEM (CONSAD RESEARCH CORP)
REGIONAL VARIABLES AT PREVIOUS YEAR: VALUE ADDED CAPITAL STOCK CONSUMPTION REGIONAL INCOME
POPULATION AND MIGRATION
GOVERNMENT EXPENDITURE
REGIONAL INDUSTRIAL INVESTMENT REQUIRED BY AIR POLLUTION ABATEMENT STRATEGY
ECONOMIC GAINS OF AIR POLLUTION CONTROL
NATIONAL INPUT/OUTPUT MODEL
REGIONAL ECONOMETRIC MODEL
REGIONAL INCOME AND CONSUMPTION
INDUSTRIAL OUTPUT INVESTMENT EMPLOYMENT CAPITAL STOCK
REGIONAL UNEMPLOYMENT LABOR FORCE
CHANGES OF REGIONAL INCOME, CONSUMPTION, VALUE ADDED, INVESTMENT AND EMPLOYMENT BY INDUSTRY, REGIONAL UNEMPLOYMENT GENERATED BY DIRECT COSTS AND BENEFITS OF EACH ABATEMENT STRATEGY

SA-1365-33

SOURCE: Woodcock, Kenneth R. A Model for Regional Air Pollution Cost/Benefit Analysis, TRW Systems Group, McLean, Virginia, May 1971

FIGURE VI-1 INTERRELATIONSHIP OF MODELS PERTAINING TO AIR QUALITY

accuracy than has probably been possible in previous studies of this type. In fact, the use of the RAPS modeling program and emission inventories to synthesize prior air pollution exposure patterns for use in epidemiological studies may be one of the more significant contributions of this RAPS experimental program.

It has been postulated that air pollution is one of the factors that causes a shift in population and the resulting changes in land use. Thus, the RAPS experimental program will be able to model air pollution concentration patterns and air pollution exposures for current periods and also to synthesize prior conditions. The results could serve as a basis for the analysis of population and land use patterns and the determination of correlations between the two sets of data. To support this correlation study, detailed survey data on population distribution, population movement, the development of new population centers, and the changes that have taken place in land use over the RAPS area will have to be obtained. If these changes can be shown to be closely correlated with air pollution exposures, then it should be possible to describe the impact of this particular situation on the structure of the regional community.

A relationship has often been claimed between air pollution or air pollution control activities and employment or unemployment within an urban or industrial area. The RAPS, especially the emission inventory and control engineering information gathering system, will provide detailed information on current air pollution and air pollution control activities within the RAPS area and probably on major changes that have occurred within the area during recent times. Detailed employment statistics are also readily available for the St. Louis area. These two sets of data should be analyzed to determine the degree to which changes in air pollution control activities or the development of air pollution problems has impacted on the labor force within the regional area. Several possible situations could be hypothesized for this relationship; the most obvious, but not necessarily the most correct, is that the imposition of air pollution control measures has caused industrial operations to close and thus to reduce the number of jobs available in a particular sector of the industry. Probably an equally likely situation is that the development of air pollution control programs within an urban area has produced a better environment for general industrial development and, through the increased attractiveness of an area for nonpolluting or pollution-sensitive industries, a regional job force has been able to expand. Both effects could obviously be going on simultaneously. By using the detailed data on community air pollution situations available through the RAPS program, it should be possible to separate these various factors as they pertain to the RAPS community.

Economic Factors

Although all air pollution impacts on a community can be interpreted in terms of economic impact, a number of conditions are recognized more clearly than others as economic factors. The RAPS program thus should be used as an experimental program through which to collect and analyze the wide range of economic impacts that can result from air pollution and air pollution control activities within a complex urban and rural regional community. An obvious task is the collection of data on the costs of inferior air quality to the general population, the commercial business sector, the industrial and manufacturing sector, and the agricultural sector. To collect these cost data it will first be necessary to develop a model of probable cost impact and then to design a practical survey technique by which data can be gathered for use in the cost model. Other segments of the RAPS program, including particularly the source emission estimation program and the air quality measurement program, can provide direct input into this cost assessment study.

As a corollary study to the assessment of the costs of inferior air quality, an assessment should be made of the costs related to control agency operations and to the application of control technology to air pollution sources. Control costs should be considered not only as direct costs, such as equipment, but indirect costs such as changes in labor utilization resulting from the introduction of new processes or the movement of industries to new locations within a community.

Because of the detailed data that will be gathered within the RAPS program on such factors as emissions, air quality, and community exposure, it seems highly likely that the RAPS program can serve as a basis for a number of special intensive studies in the economics and social area. While this utilization of the RAPS program can be expected, it is more difficult to point to a long list of intensive studies that might fall in this category. However, some of the types of studies that should be considered for this operation would include the costs and benefits of changing additives in motor fuels, the costs and benefits of detailed fuel switch operations, and the impact of specific sources, such as fluoride emissions, on neighboring community activities.

In summary, the RAPS program, although strongly oriented towards the gathering of physical data describing air pollution and its distribution within the regional area, can serve in an unparalleled way as the foundation for comprehensive analyses of the impact of air pollution and its control on the regional area. It can be argued that the success or failure of an air pollution control strategy or program must ultimately be measured by the cost and benefits that accrue to the community as a result of its application rather than simply by the observed changes in air quality that result from the modifications of emissions. In this context the projects dealing with economic and social impacts effectively closed the cycle of the RAPS research program within the regional area since the air pollution cycle begins with the source emissions, includes an atmospheric transport phase in which the pollutants are distributed across the regional area, and finally results in some economic or other impact upon the regional community.

Chapter VII

RESEARCH PLAN--TECHNOLOGY TRANSFER

Introduction

One of the major ongoing benefits of the RAPS is the fact that substantial technology will be developed that can be directly applied to air pollution control programs throughout the nation. Thus, the RAPS operation will serve as a means of upgrading local air pollution control programs and in this way will make a major contribution to the nationwide effort toward improved air quality. The need for improved technology at the local level can be seen when it is realized that most local air pollution control programs have grown primarily on the basis of local needs and with minimum reference to modern technological resources. Only in very recent years and under pressure from the Federal Air Quality Acts and increased public demands for improved air quality have local programs had the financial support that would permit them to expand their use of advanced technology in seeking solutions to air pollution problems. Thus, the RAPS with its numerous projects that can be related to local control problems comes at a very critical time in the nation's air pollution control operations; it is a time when there are increasing demands for new and improved technology and increasing resources to implement new technological programs. It is expected that the transfer of RAPS technology into the local sector will go a long way toward meeting the technological needs of air pollution control programs for new and advanced technology.

Technology Transfer Program

The RAPS staff organization has a specific office with the prescribed responsibility of directing a program to transfer technology developed for the regional area into a form whereby it can be applied to local problems. Programs within the RAPS operation that will have important impact on local operations include emission inventory techniques, air quality and meteorological measurements, data handling and analysis, the objective assessment of control strategies, and application of simulation modeling to a wide range of problems.

The RAPS program in source estimation and emission inventory should provide a basis by which local control programs can increase their effectiveness in estimating emissions within their control area and, from increased knowledge of emissions, can develop more effective control strategies and better air quality at a lower cost for the community. Emission inventory technology will be especially directed toward the development of procedures to estimate emissions over short time periods and toward improved understanding of emission factors to apply to specific sources. Improved data handling and data retrieval techniques should also result from the RAPS program and be directly translatable into local program operations.

Atmospheric monitoring networks are being required in the expanded federal program under the Clean Air Act, and the sophisticated RAPS network for both air quality measurements and meteorological measurements should contribute to increased design effectiveness and the development of more nearly optimum monitoring systems applicable to local area problems. The RAPS program will also provide means by which new technology, such as remote sensing and aircraft monitoring of local sources, can be integrated into local air monitoring programs.

Data acquisition technology including the topics already mentioned, a source inventory and air quality data, is a very important area in which improved technology can measurably improve the effectiveness of local air pollution control programs. Computer operations are becoming widespread and thus the translation of RAPS experience with automatic data acquisition, data storage, and data retrieval can provide an excellent background for upgrading local control operations in this area.

The Clean Air Act implies that local areas should use modeling technology to design implementation plans and to provide some assurance that planned control operations and control strategies will produce acceptable air quality. To date, however, suitable models that have gone through a program of verification and detailed application to air pollution control operations have not been available for the use of local agencies in their design of control strategies. The RAPS program with its heavy emphasis on model verification and subsequent model development and model revision, including air quality strategy models, can provide a major input into local air pollution control district operations because simulation modeling techniques should be available and would provide predictable degrees of accuracy and precision. The RAPS technology transfer program in the area of modeling will be able to publish descriptions of models and their performance and to supply information on the uses of these models in the formulation of implementation plans and environmental impact statements.

The design and operation of various control strategies to achieve a desired air quality within a local region is an obvious ongoing requirement for local agency operations. Within the RAPS technology transfer area, the problems of control strategy formulation will be considered as a separate topic and the various areas of the RAPS that impinge on control strategy development and control strategy testing will be integrated into one program so that this important aspect of local control operations can be presented as a single program for the use of local agencies.

The RAPS will also provide an impetus for the development of new technology through instrumentation developments and the identification of special needs for new measurement techniques and instrumental observations. Instrumentation needs that have already been identified include aerosol chemistry instruments, aircraft surveillance data acquisition packages, and meteorological sensor packages. All of these areas have obvious applications in local program areas, and, once the RAPS program has provided well-tested instrumentation designs, these procedures can be adopted by local control operations.

In summary, the RAPS technology transfer program will have the specific obligation of surveying the total RAPS operation and of translating the program carried out by RAPS into individual programs and information circulars and reports that can provide local agency programs with improved technology that has been proven by its application in the RAPS program.

Chapter VIII

OTHER AGENCY RESEARCH PROGRAMS

In developing this Prospectus, objectives and plans were developed on the basis that RAPS would be an autonomous, independent study. The possibility of collaboration with other programs that might be going on in the same area was, of course, noted, but the principle was followed that inputs and results from other programs would be supplementary rather than complementary. This principle was based upon the belief that the scope and objectives of RAPS were sufficiently important to warrant an independent, sustained, and comprehensive effort. Certainly this principle was a necessary factor in the planning phase--although the possibility could arise, when the actual instrumentation and data collection phases were begun, that some modifications and savings could be possible by taking advantage of the existing data-collection facilities. Again, the results of independent experiments would obviously be welcome in the scientific analyses and studies being conducted under RAPS. On the other hand, it is fully expected that the data collected and the results of particular experiments carried out in RAPS would be fully and readily available to other researchers working in the area, or, for that matter, to Control Agency personnel for use in their control operations.

Many reasons that prompted the selection of St. Louis for the EPA study have resulted in the selection of the St. Louis area for a number of other studies in the past, as well as currently and in prospect. The data acquired under past studies will be used in the early stages of RAPS in developing a comprehensive data bank. As to current and planned programs, it is considered most important that close cooperation and collaboration be established and maintained from the very beginning of RAPS. Specifically, the RAPS Director should maintain liaison with such programs and in particular, be at pains to ensure that any economies that can be effected by arranging for common service or shared facilities should be taken. Further, it is considered that a specific Research Task be identified that has as its objective the fullest exchange of plans, data, and results on a scientific basis. Under this Task, the Leader would be responsible for maintaining a continuing liaison with all other groups and programs carrying out research in the area, and for ensuring that there is a timely exchange of ideas, facts, and data. Thus, not only would the material aspects of cooperation be covered, but the possibly more important scientific aspects.

Of recent years it has been increasingly realized that certain problems, or groups of problems, can be usefully addressed only on an appropriately large scale, with a complex effort mounted by a number of teams of workers. RAPS itself is, of course, such an undertaking. The other major programs in progress or planned to take place in the St. Louis area are of the same type. These will be considered in turn, and discussed in terms of their relationship to RAPS.

METROMEX

METROMEX is a joint program concerned with the relationship between urban pollution and precipitation. Four major groups are involved: Illinois State Water Survey, Argonne National Laboratory, University of Chicago, and the University of Wyoming. The project completed its first season in the summer of 1971. The leader of the project is Mr. Stanley Changnon of Illinois State Water Survey, and funding is largely provided by NSF, AEC, and the State of Illinois. The Illinois State Water Survey element has the following goals: (1) to explore the magnitude and distribution of urban precipitation, (2) to study the temporal and spatial variation of artificial tracers, and (3) to study local severe storms. The primary monitoring entails a network of 220 weighing bucket rain gauges within a 25-mile radius of St. Louis, half of which are to be in operation throughout the year. Two weather radars are operated during the experimental period and data were also collected from six recording wind stations located around the 25-mile circle. Analyses were also made of drop size distributions, the rain collected at 70 stations was analyzed for trace indicators, and a number of other records were made including temperature and humidity at seven stations.

The Argonne National Laboratory element was concerned with the mechanical and thermal effects of the urban core upon the wind distribution over the area. The primary tool was a network of nine PIBAL (pilot balloon) stations from which wind profiles were measured simultaneously at 20-minute intervals by theodolite observations. Work was also carried out on the analysis of rain water to measure the presence of some 10 trace elements. The University of Chicago element was concerned with the cloud physics of the effect of the city on precipitation: (1) as shown by variations in nuclei population, (2) size distribution of cloud droplets, (3) precipitation mapping by radar, and (4) delineating the urban cloud plume. Observations were made by an additional ground-based radar and from an aircraft, which also had a radar, but whose main role was in situ sampling of the cloud and aerosol.

The University of Wyoming element had four objectives: (1) a study of the mesoscale circulation in conjunction with their modeling efforts, (2) a study of the Aitken and ice nuclei in urban areas, and (3) study of the effects of urban areas on cloud growth. Most of the observations were carried out by in situ sampling from an aircraft in flight, but PIBAL observations were made at four stations and a series of radiosonde measurements was also made.

It will be seen that the objectives of METROMEX coincide to a large extent with those of Task 204 in the RAPS program.

NCAR Fate of Pollutants Study (FAPS)

During the past season's work on METROMEX, NCAR participated by carrying out what is intended to be the first year's work on a more extensive, independent study entitled, "Fate of Pollutants Study." Its objectives are to determine the extent to which various pollutants are removed from the air by nonmeteorological processes, such as chemical reaction, absorption by plants, adsorption on surfaces and bacterial action in the soil, and are thus prevented from contributing to regional and world-wide atmospheric pollution. The NCAR activities at the time of the METROMEX work were designed to answer certain questions related to the experimental plan of the major FAPS study. These were: (1) Can the plume position downwind of the city be forecast with sufficient accuracy to allow stations to be placed in the plume? (2) At a distance of 120 km, is there sufficient contrast between concentration in the plume and the background concentration? Both these questions were answered in the affirmative. In addition, they explored whether additional sources were present downwind of the city to cause complications in the interpretation of the data. Some sources of SO_2 were found to exist but could be identified and allowed for. (Concentrations of NO_2, however, appeared to fall to the background level within 50 km or so of the city.)

Longer-term plans of NCAR include sampling along two arcs, both subtending an angle of 80° from the center of the city and located 80 and 120 km respectively, downwind. A variety of pollutants will be measured at ground stations and from the air along these areas. Interpretation of the data will be undertaken on three bases:

(1) Model concentration comparison--relating measurements with model computations for reactive species, validated against unreactive species such as CO_2

(2) Flux through two planes--in which a mass balance of various pollutants can be established

(3) Ratios of pollutants--in which the relative concentrations of various species are compared to determine the change and rate of change of concentration of various contaminants.

Thus it will be seen that the NCAR program, which is scheduled for its major session in the late summer of 1973, has very similar objectives and approaches as the RAPS Task 205, and it is to be hoped that the results of both experiments can be mutually supporting and beneficial.

NOAA's MESOMEX

A third major study in prospect is that planned by NOAA. The objectives of this study are a better understanding and prediction capability (preferably objective) of atmospheric conditions in the boundary layer on the mesoscale (250 by 250 km or so). Some initial experiments have in fact been carried out in the Oklahoma City area last summer (1971), but it is understood that the St. Louis area is being considered for this project.

It would seem that this program would relate very closely to RAPS plans for modeling the meteorological conditions of the boundary layer, which have been described in the Research Plan Task 101.

Additionally recognizing the value of carrying out individual experiments in areas in which minor observational programs are being carried out, a number of separate research projects are in progress or have been planned in the St. Louis area.

Finally, relevant research programs outside the St. Louis area should be noted by the Task Leader of Task 400, and every effort made to ensure a free flow of information in and out of the RAPS project. A particularly important series of major experiments is about to be carried out by various research teams under the sponsorship of the State of California and the Coordinating Research Council in various parts of California. Since these experiments will be a major source of information on photochemical effects, particularly those related to smog formation from automotive products, their results should be carefully considered in the RAPS study for, in the St. Louis area, such effects will not be highly developed.

In conclusion, it appears that there is a considerable potential for deriving benefits for the RAPS program from other ongoing programs, and vice versa. Certainly, care must be taken to ensure that there is no wasteful duplication of effort, although some overlap is probable and not wholly undesirable. Regardless of other efforts in the same and similar areas, it would be important to maintain a well-focused RAPS effort in its own right for two reasons: (1) only by so doing will it be possible to ensure a closely meshed, well-integrated contribution to the overall RAPS program. (Many of the other proposed studies are planned to operate on the basis of short-term intensive experiments, rather than on an extended basis.) (2) It is emphasized that RAPS objectives are directed specifically towards a different goal. In the other programs noted, the emphasis is mainly on the scientific aspects of the problems attacked, or upon operational problems quite different from those of RAPS. RAPS is concerned with the development of improved air pollution control strategies, and this concept must dominate all its research tasks.

Appendix

SCHEDULES AND TASK SPECIFICATIONS FOR THE RESEARCH PLAN

SCHEDULES AND TASK SPECIFICATIONS FOR THE RESEARCH PLAN

Introduction

The following schedules and specifications show how the Research Plan would be accomplished within the proposed 5-year period. They follow a structure reflecting the principal objectives of the study. Thus, each principal objective is approached within principal tasks numbered 100, 200, 300, and 400, respectively. Separate research tasks are assigned numbers within the 100, 200, 300, and 400 series, and a further division into subtasks is provided by adding numbers after the decimal point. For example, Task 101 Boundary Layer Meteorology is a task within the 100 series which is concerned with Model Verification. (Task No. 100). Task 101.1 Area Climatology is an element of Task 101. A task specification is given for each task, which states the objective and purposes and scope of the research element concerned. It is intended that professional responsibility for each task be assigned to an individual, who would be charged with accomplishing his task in terms of its stated objective. The Principal Tasks 100, 200, 300, and 400 in fact would be carried out by the senior personnel of the project as part of their assigned responsibilities. However, the leaders of the subordinate tasks and subtasks (e.g., 101 and 101.1) would be engaged in the day-to-day accomplishment of the work specified, each task leader of a separate task (e.g., 101) being responsible for the product of the subtask leaders (e.g., 101.1) and responsible to the principal task leader (e.g., 100).

Specifications for these tasks follow. Also presented are schedules showing the general timing of the activity within the five-year period. In both cases, the amount of effort that has been used as a basis for planning and costing is indicated in terms of man-years of professional and subprofessional effort.

Preceding page blank

100 MODEL VERIFICATION

	1972	1973	1974	1975	1976	1977
101 Boundary Layer Meteorology (2.5 p)	xx	xxxx	xxxx	xxxx	xxxx	xx
101.1 Area climatology (.5 p, .5 s)	xx	xx..	....	....	....	..
101.2 Prior diffusion data (.5 p, .5 s)	xx	xx..	....	....	....	..
101.3 Compilation and analysis of upper air data (2.1 p, 2.1 s)	..	.xxx	xxxx	xxxx	xxxx	xx
101.4 Compilation and analysis of near-surface data (2.1 p, 2.1 s)	..	.xxx	xxxx	xxxx	xxxx	xx
101.5 Balloon-tracking experiment (.5 p, 1.5 s)	..	....	.xx.	....	....	..
101.6 Diffusion tracer experiments (.5 p, 1.5 s)	..	....	....	.xx.	....	..
101.7 Weather satellite applications (1.5 p, .75 s)	..	....	....	xxxx	xx..	..
101.8 Forecast models (2 p, 2 s)	..	....	....	..xx	xxxx	xx

Total effort 12.25 man-years--professional (p)
11.00 man-years--subprofessional (s)

100 MODEL VERIFICATION (Continued)

		1972	1973	1974	1975	1976	1977
102	**Emission Inventory** (5 p)	xx	xxxx	xxxx	xxxx	xxxx	xx
102.1	Emission inventory design (1 p, 1 s)	.x	x...	....	....	....	..
102.2	Collection of emission data for stationary sources (5.25 p, 5.25 s)	..	xxxx	xxxx	(xxxx	xxxx	xx)
102.3	Collection of emission data for mobile sources (.5 p, 5.25 s)	..	.xx(x	xxxx	xxxx	xxxx	xx)
102.4	Emission model for stationary sources (1 p, 1 s)	..	.xxx	xxxx	x...	....	..
102.5	Emission model for mobile sources (1 p, 1 s)	..	..xx	xxxx	xx..	....	..
102.6	Emission source test (6 p, 6 s)	..	..xx	xxxx	xx(xx	xxxx	xx)
102.7	Analysis of status of source controls (1.25 p)	..	.xxx	....	....	....	..
102.8	Emissions inventory of agricultural data sources (.25 p, .25 s)	..	.xx.	....	....	....	..

Total effort 20.25 man-years--professional
19.75 man-years--subprofessional

100 MODEL VERIFICATION (Continued)

	1972	1973	1974	1975	1976	1977
103 Air Quality Measurement (4.25 p)	xx	xxxx	xxxx	xxxx	xxxx	xx
103.1 Data base (.25 p)	.x	x...	....	....	....	..
103.2 Air quality data from local agencies (1.18 p, 4.75 s)	.x	xxxx	xxxx	xxxx	xxxx	xx
103.3 Analysis of air quality and meteorological data acquired by RAPS (4.5 p, 8.5 s)	.x	xxxx	xxxx	xxxx	xxxx	xx
103.4 Analysis of air quality data acquired by aircraft (1.06 p, 2.1 s)	..	.(xxx	xxxx	xxxx	xxxx	xx)
103.5 Fine scale spatial variation of air quality (.75 p)	..	xxxx	xx..	....	....	..

Total effort 12 man-years--professional
15.35 man years--subprofessional

100 MODEL VERIFICATION (Concluded)

		1972	1973	1974	1975	1976	1977
104	Model Calculation and Verification (2.5 p)	xx	xxxx	xxxx	xxxx	xxxx	xx
104.1	Evaluation of selected models (30 p, 70 s)	xx	xxxx	xxxx	xxxx	xxxx	xx
104.2	Model modification and improvement (2 p, 2 s)	..	..xx	xxxx	xxxx	xxxx	xx
104.3	Methodology for determining model accuracy (1.88 p)	..	...x	xxxx	xxxx	xxxx	xx
104.4	On-site computation and data display (2 p)	..	..xx	xxxx	xxxx	xxxx	xx

Total effort 38.35 man-years--professional
72 man-years--subprofessional

200 ATMOSPHERIC, CHEMICAL, AND BIOLOGICAL PROCESSES

		1972	1973	1974	1975	1976	1977
201 Gaseous Chemical Processes (5 p shared with 202)		xx	xxxx	xxxx	xxxx	xxxx	xx
201.1	Hydrocarbon analyses and monitoring (2.06 p, 5.63 s)	..	....	xxxx	xxxx	xxxx	..
201.2	Development of hydrocarbon classifier instrumentation (1.38 p)	xx	xxxx	....	....	....	..
201.3	Total aldehyde-formaldehyde monitoring program (.19 p, .31 s)	..	....	xxxx	xxxx	xxxx	..
201.4	Determination of peroxyacetyl nitrate (.44 p, .75 s)	..	....	....	xxxx	xxxx	..
201.5	Ammonia monitoring program (1 p, 3.12 s)	..	...x	xxxx	xxxx	xxxx	xx
201.6	CO, SO_2, and NO_2 mass flux measurements (.81 p, .88 s)	..	....	..xx	xxxx	xxxx	xx
201.7	Origin of atmospheric CO (106 p, .75 s)	..	....	....	..xx	xxxx	xx
201.8	Atmospheric odor identification (.75 p, 1 s)	..	....	....	....	...x	xx

Total effort 11.69 man-years--professional
12.45 man-years--subprofessional

200 ATMOSPHERIC, CHEMICAL, AND BIOLOGICAL PROCESSES (Continued)

		1972	1973	1974	1975	1976	1977
202 Atmospheric Aerosol Processes	(Shared with 201)	xx	xxxx	xxxx	xxxx	xxxx	xx
202.1	Determination of total nitrate in aerosol samples (.31 p, 1.5 s)	..	....	xxxx	....	....	..
202.2	Determination of total sulfate in aerosol samples (.31 p, 1.38 s)	..	....	xxxx	....	....	..
202.3	Determination of aerosol size-distribution (.69 p, 2.75 s)	..	....	xxxx	xx..	....	..
202.4	The NO_2NaCl reaction in aerosol (.25 p, .13 s)	..	....	.xxx	xxxx	xxxx	xx
202.5	Isotope ratios of sulfate aerosols (.56 p, .75 s)	..	....	....	..xx	xxxx	..
202.6	Organic compounds in particulate material (.75 p, .88 s)	..	....	....	xxxx	xx..	..
202.7	Experimental measurements of deposition velocity (.56 p, 2.5 s)	..	....	....	....	xxxx	xx

Total effort 3.44 man-years--professional
9.87 man-years--subprofessional

200 ATMOSPHERIC, CHEMICAL, AND BIOLOGICAL PROCESSES (Continued)

	1972	1973	1974	1975	1976	1977
203 Other Pollutant Related Processes	xx	xxxx	xxxx	xxxx	xxxx	xx
203.1 Radiation balance modification (.69 p, 1 s)	..	..xx	xxxx	xxxx	....	..
203.2 Visibility reduction in urban and rural areas (.69 p, 1 s)	..	....	....	xxxx	xxxx	..
203.3 Transport of atmospheric odors (.25 p, .25 s)	..	....	....	....	..xx	xx
203.4 Trace metals and toxic trace materials (.69 p, 2.5 s)	..	....	....	xxxx	xxxx	..
203.5 Agricultural chemical distribution (.25 p, .75 s)	..	....	....	..xx	xxxx	xx
203.6 Natural sources of air polluted compounds (1.13 p, .19 s)	..	....	xxxx	xxxx	xxxx	..

Total effort 3.69 man-years--professional
5.69 man-years--subprofessional

200 ATMOSPHERIC, CHEMICAL, AND BIOLOGICAL PROCESSES (Continued)

		1972	1973	1974	1975	1976	1977
204	Atmospheric Scavenging by Precipitation						
204.1	Instrument development for pH and chemical sampling (.50 p, .5 s)	xx	xxxx	....	....	....	..
204.2	Rainfall pH measurement (.53 p)	..	....	xxxx	xxxx	xxxx	xx
204.3	Measurements of rainfall chemistry (.88 p, 5.25 s)	..	....	.xxx	xxxx	xxxx	xx

Total effort 1.90 man-years--professional
5.75 man-years--subprofessional

		1972	1973	1974	1975	1976	1977
205	Air Pollutant Scavenging by the Biosphere						
205.1	Chemical content of vegetation (.5 p, 2 s)	..	....	.xx.	..xx	xxxx	..
205.2	Atmospheric pollutant concentrations related to vegetation absorption (.5 p, 1.5 s)	..	....	....	..xx	xxxx	xx

Total effort 1 man-year--professional
3.5 man year--subprofessional

200 ATMOSPHERIC, CHEMICAL, AND BIOLOGICAL PROCESSES (Concluded)

		1972	1973	1974	1975	1976	1977
206 Atmospheric Processes (1.5 p, 3 s)							
206.1	Source factors in pollutant dispersal (.5 p)	..	..xx	xxxx	xxxx	xx..	..
206.2	Terrain and surface roughness effects (.38 p)	..	....	.xxx	....	....	..
206.3	Extraregional and synoptic scale circulation (.75 p)	..	....	...x	xxxx	xx..	..

Total effort 3.12 man-years--professional
3 man-years--subprofessional

300 HUMAN, SOCIAL, AND ECONOMIC FACTORS

	1972	1973	1974	1975	1976	1977
301 Human and Social Factors						
301.1 Data on epidemiology and health effects (4.5 p, 4.5 s)						
301.2 Data on population and land-use characteristics	Continuing low scale effort					
301.3 Data on labor force utilization						

Total effort 4.5 man-year--professional
4.5 man-year--subprofessional

	1972	1973	1974	1975	1976	1977
302 Economic Factors						
302.1 Costs of inferior air quality to industrial and general population (4.5 p, 4.5 s)						
302.2 Costs of control strategies	Continuing low scale effort					
303.3 Data collection surveys of specific effects						

Total effort 4.5 man-years--professional
4.5 man-years--subprofessional

400 TRANSFER OF RAPS TECHNOLOGY FOR CONTROL AGENCY APPLICATIONS AND THE FORMULATION OF CONTROL STRATEGIES

	1972	1973	1974	1975	1976	1977
401 Source Inventory Procedures (1 p, 1 s)						
401.1 Techniques for making source inventory procedures						
401.2 Techniques for inventory storage and retrieval						
	..	....	....	xx..	....	xx
401.3 Techniques for updating the source inventory						
401.4 Relating source inventory to control strategy						

Total effort 1 man-year--professional
1 man-year-subprofessional

400 TRANSFER OF RAPS TECHNOLOGY FOR CONTROL AGENCY APPLICATIONS AND THE FORMULATION OF CONTROL STRATEGIES (Continued)

402 Atmospheric Monitoring

		1972	1973	1974	1975	1976	1977
402.1	Basic network principles (.5 p)	..	....	..xx	....	....	xx
402.2	Criteria for organization and maintenance of observational networks (.5 p)	..	....	xx..	....	....	xx
402.3	Station siting and instrument exposure criteria (.25 p)	..	....	....	xxxx	xxxx	xx
402.4	Methodology for modernization of monitoring networks (.25 p)	..	....	xx..	....	....	xx
402.5	Evaluation of new aircraft measurement techniques (1.63 p)	..	..xx	(xxxx	xxxx	xxxx	xx)
402.6	Evaluation of remote measuring techniques (2.75 p, 3 s)	..	..xx	(xx	xxxx	xxxx	xx)

Total effort 5.89 man-years--professional
3 man-years-subprofessional

400 TRANSFER OF RAPS TECHNOLOGY FOR CONTROL AGENCY APPLICATIONS AND THE FORMULATION OF CONTROL STRATEGIES (Continued)

403 Data Handling

		1972	1973	1974	1975	1976	1977
403.1	Optimize techniques for data acquisition, storage, and retrieval (.5 p)	..	....	..xx	....	....	xx

Total effort .5 man-year--professional

404 Modeling Technology

		1972	1973	1974	1975	1976	1977
404.1	Significance of modeling to the formation of control strategies and their implementation (2.25 p)	xx	xxxx	xxxx	xxxx	xxxx	xx
404.2	Implementation Plan applications (.75 p, .75 s)	..	....	xx..	....	..xx	xx
404.3	Environmental Impact Statement applications (.75 p, .75 s)	..	....	xx..	....	..xx	xx
404.4	Methodology for assessing model validity in control agency operations (.75 p, .75 s)						

Total effort 4.5 man-years--professional
2.25 man-years--subprofessional

400 TRANSFER OF RAPS TECHNOLOGY FOR CONTROL AGENCY APPLICATIONS AND THE FORMULATION OF CONTROL STRATEGIES (Concluded)

	1972	1973	1974	1975	1976	1977
405 Other Significant Factors in Control Strategy Formulation						
405.1 Liaison and interaction with other environmental research programs						
405.2 Techniques of assessing social and economic factors						
405.3 Methodology of assessing operational costs of control strategies	Continuing low scale effort					
405.4 Methodology of assessing resultant costs of control strategies						
405.5 Institutional aspects						

Total effort	4.5 man-year--professional 4.5 man-year--subprofessional

100 Model Verification

Objective

The objective is to test, evaluate, and verify the capability of mathematical simulation models to describe and predict the transport, diffusion, and concentration of both inert and reactive pollutants over a regional area.

Purpose and Scope

The purpose of this series is to investigate the performance of a series of selected mathematical simulation models in circumstances such that as many variables as possible are known. In this way the validity of the model can be determined, or its deficiencies identified, and methods of measuring input or verification data can be optimized for use in subsequent operational applications. The program will investigate emission source assessment, the modeling of atmospheric physical and chemical factors, and the methods of verifying and evaluating the operation of the models.

The approach follows two major lines. The first approach is development of the most complete description possible of the meteorological conditions, emissions, and air quality. The second approach applies the appropriate input data to a series of selected models and examines their product against observed data.

The selection of models will follow an evolutionary program, starting with the models ready for immediate testing. Where possible, competing models will be applied to the same data sets. Subsequent tests will be made of improvements of such models on an iterative basis, or in combinations of the most successful features of such models. Major emphasis will be on models relating to the regional scale, but subscale models will be treated in time. First priority will be given to the development of useful tools for reviewing and assessing Implementation Plans. The similar need for Environmental Impact Statements will be given concurrent but subordinate attention. Subsequently, the aim will be to provide fully tested optimum models, the performance of which can be objectively assessed, at each scale for each main class of pollutants.

Consideration will also be given to the role of modeling in connection with air pollution episode prediction.

101 Boundary Layer Meteorology Program

Objective

The objective is to collect meteorological data on the parameters that affect the dispersion of atmospheric pollutants in the St. Louis region.

Purpose and Scope

The purpose of this task is to:

- Provide the meteorological data that are required as inputs to air quality simulation models.
- Obtain data with which to evaluate model computations of meteorological parameters.
- Provide supplemental measurements of meteorological parameters for study of fundamental meteorological processes for subsequent use in the revision of various model components.

The program will commence with the collection of historical climatological and experimental diffusion data for the St. Louis area. Later, data from the various routine RAPS meteorological facilities will be assembled and compiled in a meaningful format using standard metric units; the routine facilities include the basic surface network and the upper air (aircraft and balloon) systems. The program will also include collection and compilation of data from special research programs.

101.1 Area Climatology

Objective

The objective of this subtask is to determine the nature of the climatology of the St. Louis region as it is related to the dispersion and transformation of atmospheric pollutants.

Purpose and Scope

The purpose of this subtask is to assemble climatological data from the St. Louis region to aid in station siting, as well as in the direction and scope of atmospheric research programs that are aimed at the study of dynamic, kinematic, energetic, and chemical processes. Also, the research will provide data for input to, and the evaluation of, climatological air quality simulation models.

101.2 Prior Diffusion Data

Objective

The objective of this subtask is to determine the probable nature of atmospheric diffusion properties in St. Louis on the basis of earlier diffusion experiments in this and other similar regions.

Purpose and Scope

The purpose of the research is to collect results of prior diffusion experiments to assist in the planning of special research studies within RAPS, as well as in the number and spacing of surface-based monitoring stations. These data will be summarized through stratification by the relevant physical parameters and used for the planning of the scope and direction of diffusion studies in St. Louis.

101.3 Compilation and Analysis of Upper Air Data

Objective

The objective of this subtask is to provide meteorological data on the initial and boundary conditions of the three-dimensional structure of the planetary boundary layer.

Purpose and Scope

The work in this subtask will provide these data for input to, and the subsequent evaluation of, regional air quality simulation models. These data will be obtained through the use of balloon-tracking, helicopter, and airplane systems and therefore will also provide flexibility in the support of special research programs, e.g., radiation, tracer, transformation, and long range dispersion studies. The data from the routine upper-air network will also be used for the development of an extensive climatology of regional boundary layer meteorology.

101.4 Compilation and Analysis of Near-Surface Data

Objective

The objective of this subtask is to provide relatively detailed and comprehensive micrometeorological data on the three-dimensional structure of the atmospheric surface layer on the regional scale.

Purpose and Scope

Research will be conducted to obtain these micrometeorological data for input to air quality simulation models and for their subsequent verification. The data will also be used to study the effects of topographic, anthropogenic, and pollutant features together with mesoscale weather types, on the dynamics of low-level atmospheric motions and the feedback on the distribution of contaminants. As such, this program will support many of the special research experiments as well.

101.5 Balloon-Tracking Experiment

Objective

The objective of this experiment is to provide information on Lagrangian atmospheric characteristics, as well as on their spatial distribution.

Purpose and Scope

Purpose of the experiment is to provide these data for studies seeking to evaluate the spatial resolution required in the specification of upper-level meteorological fields (e.g., wind, temperature, humidity) on the mesoscale in consideration of the distribution of surface features (e.g., roughness, drainage, soil type). More specifically, these data will be available for use in studies of long range dispersion, evaluation of topographic effects, city-induced changes in wind structure, urban heat island, and urban-rural radiation difference. In summary, the purpose is to provide input to the detailed study of upper-level atmospheric processes and their incorporation in air quality simulation models.

101.6 Diffusion Tracer Experiments

Objective

The objective of these experiments is to provide information on the diffusion of a known trace element under a variety of conditions of meteorology, topography, and source characteristics.

Purpose and Scope

The experiments will be conducted to provide the necessary data for studies of the effects of atmospheric, topographic, and source characteristics on the diffusion of plumes from isolated point sources as well as trace element(s) that can be released under controlled conditions of source rate, duration, height, and type. These data also will provide a selected base for the evaluation and subsequent verification of certain types of air quality simulation models.

101.7 Weather Satellite Applications

Objective

The objective of this subtask is to assess the value of meteorological data obtained by satellite to the RAPS program and for use in air quality monitoring in general.

Purpose and Scope

Meteorological satellites offer the potential of being able to provide meteorological data of the type required in air pollution programs on a routine basis and at a minimal cost. This program should seek to define objectively the specific advantages and limitations of these data to the special and routine studies embodied in the RAPS program. If the evaluation shows the desirability of obtaining these data in St. Louis, then the next task will be the acquisition of the data and subsequent cataloging as required by the specific purposes or experiments these data will serve.

101.8 Forecast Models

Objective

The objective of this subtask is the incorporation of routine, weather prediction models operated by other agencies into the RAPS program.

Purpose and Scope

Numerical weather prediction models are currently in operational use by three agencies in the United States: NOAA (National Weather Service), the Navy (Fleet Numerical Weather Facility), and the Air Force (Global Weather Central). Of these groups, only the Air Force routinely operates a mesoscale, boundary layer model. The purpose of this program is to assess the advantages of incorporating the routinely available boundary layer forecast into the RAPS program. These forecasts may provide the mesoscale boundary conditions required by some prognostic air quality models or they may even provide the necessary prognostic meteorological field. Additionally, the evaluation of the applicability of these forecasts should consider their benefit in experimental planning. If these data can be efficiently used within RAPS, the assessment program must also consider problems of data acquisition, format and display.

102 Emission Inventory

Objective

The objective of this task is to develop and maintain a comprehensive source inventory.

Purpose and Scope

Numerous components of the Research Plan will require a complete inventory of all emission sources in the study area. This will include both fixed and mobile sources of all major pollutants and perhaps selected minor materials. Emission levels must be described for selected times ranging from yearly to hourly intervals. The inventory should serve in the validation of models, analysis of control strategies, and the investigation of air quality impacts on the human, social, and economic systems of the area.

102.1 Emission Inventory Design

Objective

The objective of this subtask is to develop a detailed method for taking an inventory of the emissions from both stationary and mobile sources.

Purpose and Scope

Emission inventories must be designed to serve a variety of uses, including input data for model verification programs, studies of control strategies, and impact analyses. The development of emission inventories can be an extremely costly enterprise requiring extended periods of time. For these two reasons, a specific task is required to design procedures for an emission inventory before full scale field surveys.

The task should first provide for the review of previous inventories to identify the method used to classify sources by type, location, emission levels over time, and other characteristics. The desirability of incorporating these data into the Regional Study emission inventory should be assessed.

Procedures should then be developed for obtaining data describing all classes of sources, such as point, area, and mobile. Questionnaires should be designed to cover certain classes of sources, whereas procedures may be designed for physical measurement of other sources. In the case of vehicular emissions, procedures may be required to determine traffic flows to be combined with the characteristics of the sources themselves.

Finally, methods for computer storage and retrieval of the emission inventory data will require development.

102.2 Collection of Emission Data for Stationary Sources

Objective

The objective of this subtask is to collect emission data from both point and area sources and to maintain these data on a continuing basis.

Purpose and Scope

The purpose of this effort is to identify pollution sources in terms of the chemical and physical characteristics of their emissions, the magnitude over time of the emissions, their location, and all other descriptors as may be required for purposes of model evaluation, control strategy development, impact studies, and the like.

The collection of emission data initially will emphasize the predominant pollutants, such as SO_2, CO, NO_x, and particulates, originating primarily from combustion sources but including the larger noncombustion sources. The general sequence of collection will be derived from existing emission inventories reviewed in 102.1 to eliminate duplication of valid current data. Data will be collected by questionnaire and on-site visits. As this effort is concluded, the survey of sources of lower emissions, such as industrial processes, will be initiated. Pollutants in addition to those listed above will also be included.

This task will continue at a relatively low level through the Regional Study for continual revision of the inventory as source operations change or are controlled.

102.3 Collection of Emission Data for Mobile Sources

Objective

The objective of this subtask is to collect emission data from mobile sources, including motor vehicles and aircraft.

Purpose and Scope

The purpose of this effort is to determine the flow pattern of motor vehicles, aircraft, and other mobile sources of interest as a function of time and by their emission characteristics. The mobile units will be characterized by selected characteristics, including size, fuel, engine type, and similar features and emission factors assigned to each.

Flow patterns of mobile sources in the region will first be defined. The definition will be developed by review of available motor vehicle traffic studies in the region, airport schedules, and similar sources. Since the street and highwaypattern in the area is undergoing significant changes, field surveys may be required for the more important thoroughfares not included in existing data.

This effort should continue throughout the study and be revised as vehicle emission control techniques and movement patterns change.

102.4 Emission Model for Stationary Sources

Objective

The objective of this subtask is to develop a model for predicting emissions from stationary sources over specified time intervals.

Purpose and Scope

The purpose of this task is to develop a set of models to predict emissions from stationary sources under specified conditions for periods ranging from one hour to one year. Baseline emission estimates for specific conditions of the source first will be developed. These conditions perhaps can be defined as the average or typical conditions of the source. Preparation of adjustment factors for correcting the baseline emissions for other conditions of the source will follow. These adjustment factors will include numerous variables, including atmospheric temperature, time of day, season of year, and shifts in the type of fuel used. Additional factors, especially important for industrial plants, will include variations in shift levels and resulting production rates, together with changes in product mix. The emission model undoubtedly will be sufficiently complex to require the use of computer techniques in its development and operation.

102.5 Emission Model for Mobile Sources

Objective

The objective of this subtask is to develop a model for predicting emissions from mobile sources over specified time intervals.

Purpose and Scope

The purpose of this task essentially parallels that of 102.4. On the basis of the survey of 102.2, selected models will be developed covering motor vehicle flows, aircraft movements, and perhaps railroad and ship activities. These movement patterns will be combined with the emission factors to develop estimates of emissions for specified periods ranging from one hour to one year. The complexity of the model undoubtedly will be such that a computer will be required.

102.6 Emission Source Test

Objective

The objective of this subtask is to conduct field measurements of emissions from stationary sources and develop emission factors.

Purpose and Scope

Although emission factors for many sources can be adequately estimated by comparison with similar sources or by other methods, the more important combustion and noncombustion sources will require field measurements to determine reliable emission factors. Measurements will be made under a range of source conditions, such as level of production and electric power output and correlated with emission characteristics. These analyses should produce reliable baseline emissions and adjustment factors for incorporation into the emission model developed in 102.4.

102.7 Analysis of Status of Source Controls

Objective

The objective of this analysis is to determine the degree of emission control in effect.

Purpose and Scope

The stationary source inventory should reveal the extent to which emission control equipment and operational procedures are used in the area along with measures of their effectiveness and perhaps cost. These data will be used to determine efficiency of these control measures and in conjunction with any implementation plans.

102.8 Emissions Inventory of Agricultural Sources

Objective

The objective of this subtask is to estimate the emissions from agricultural operations within the St. Louis region.

Purpose and Scope

A wide variety of materials are emitted to the atmosphere as a result of agricultural operations. These include fungicides, insecticides, and herbicides; emissions of contaminants from burning agricultural waste products; and dust from plowing, harrowing, and the like. Emission factors covering these and perhaps other materials will be required as part of the total emission inventory of the region in terms of both baseline levels and adjustment factors.

103 Air Quality Measurement

Objective

The objective of this task is to provide as complete and detailed a description as possible of the distribution (both in space and time) and concentration of air pollutants in the St. Louis region.

Purpose and Scope

The purpose of this task is to provide the data on pollutant distribution necessary for the verification of simulation models of various scales (in space and time) and for various types of pollutant (i.e., both reactive and nonreactive) and various types of source.

Data will be obtained from past records, from routine measurements already being made in the St. Louis area, and from the special measurement networks of the RAPS facility.

103.1 Data Base

Objective

The objective of this subtask is to provide a data base of relevant information concerning pollutant concentrations measured in past air quality surveys of the St. Louis area that can be integrated into the RAPS and used in developing methodologies for using such data in other areas.

Purpose and Scope

The purpose of this subtask is to take advantage of the data handling and analysis capabilities of the RAPS organization to correlate historical pollution data with RAPS network data to delineate changes and trends in pollutant concentrations and to indicate disparities due to measurement techniques, site locations, source locations, and the like. Also, this subtask will determine the effectiveness of control and abatement procedures.

103.2 Air Quality Data from Local Agencies

Objective

The objective of this subtask is to collect and evaluate air quality measurement data acquired by the local agencies, Air Pollution Control, City of St. Louis, and Air Pollution Control, St. Louis County, and to integrate agency measurements into RAPS program and initiate agency access to RAPS data.

Purpose and Scope

Purposes of this subtask are to:

- Assemble and correlate data from the existing local monitoring networks with that obtained by RAPS with respect to site location, measurement techniques, and data handling so as to take advantage of the data handling and analysis capability of RAPS.

- Evaluate the effectiveness of a modest network to obtain measurements representative of the area of interest.

103.3 Analysis of Air Quality and Meteorological Data Acquired by RAPS

Objective

The objective of this subtask is to develop computer software for compilation, retrieval, and analysis of the data acquired by the RAPS monitoring network.

Purpose and Scope

This effort will be to take advantage of the data handling and analysis capability of a RAPS organization to provide data tabulation, measurement correlations, and continuously updated analysis techniques. Analysis display techniques, such as isopleth maps and displays to simplify data interpretation, also will be developed.

103.4 Analysis of Air Quality Data Acquired by Aircraft

Objective

The objective of this subtask is to develop computer software for compilation, retrieval, and analysis of the air quality data acquired by the airborne laboratory.

Purpose and Scope

This subtask will take advantage of the data handling and analysis capability of a RAPS organization to provide data tabulation, aircraft to network measurement correlation, horizontal and vertical profile analysis and display, flight pattern mapping, concentration mapping, and, where sufficient data are obtained, isopleth mapping.

103.5 Fine Scale Spatial Variation of Air Quality

Objective

The objective of this subtask is to ascertain the subgrid scale spatial variability of air quality in the region as a function of emissions, meteorology, and topography.

Purpose and Scope

This subtask will assess, through a series of field measurement programs, the fine-scale variability of air quality throughout the region. The variability should be assessed over areas corresponding to the model resolution that is required so that the representativeness of measurements and predictions of air quality can be evaluated.

These assessments will be vital to the intelligent evaluation and verification of simulation models, as well as toward an understanding of the influence of distributions of emission, meteorological, and topographical factors on the level and variance of air quality.

104 Model Calculation and Verification

Objective

The objective of this task is to operate, evaluate, and modify a series of air quality simulation models and identify the models that are most suitable for use in formulating air pollution control strategies.

Purpose and Scope

Purposes of this task are to:

- Provide evidence and information as to the effectiveness of a series of selected candidate models so that optimum models can be selected for each aspect of air pollution control.

- Develop techniques of categorizing models in terms of their applicability in respect to both the temporal and spatial resolution and the type or class of pollutants for which they are appropriate.

- Develop techniques of comparing and assessing the relative accuracy and precision of models.

- Provide a local capability for carrying out computations and investigating the way in which submodels may be developed or major models improved.

104.1 Evaluation of Selected Models

Objective

The objective of this subtask is to operate, test, and evaluate a series of air quality simulation models.

Purpose and Scope

The purpose of this subtask is to carry out actual modeling procedures using input data in various degrees of sophistication and completeness with a series of candidate models. The products of these procedures then will be confronted with air quality measurements and the validity of the model assessed, using methodologies developed in parallel in Task 104.3.

Models will be categorized in terms of their applicability for various types of pollutants and for various spatial and temporal resolutions. Their fitness for use for air pollution control operations and the formulation of improved strategies (short term episodes and longer term occurrences) will be assessed by evaluating their: (1) mathematical integrity, (2) nature and extent of input data, and (3) utility of the output data format of the model, in addition to their accuracy and precision.

104.2 Model Modification and Improvement

Objective

The objective of this subtask is to develop the best available forms of candidate models or of combinations of their subelements.

Purpose and Scope

The purpose of the subtask is to provide an opportunity to experiment with and investigate optimum combinations of elements of candidate models in the light of their use in the test and evaluation phase of this study, i.e., in Task 101.4. (In practice, this task may well be incorporated with Task 104.1. It is treated separately here for planning and costing purposes.)

104.3 Methodology for Determining Accuracy

Objective

The objective of this subtask is to provide a methodology by which the accuracy of various models may be measured and assessed.

Purpose and Scope

The purposes of the subtask are to:

- Develop objective techniques to facilitate the comparison of models, where rival candidates exist.

- Develop ways of evaluating and describing the accuracy and precision of any proposed model in terms of its potential use in the formulation of control strategies (episode and long term). In this way, the sensitivity of models to the degree of completeness and sophistication of their input data can be assessed. Additionally, the usefulness of the models' output as determined from the user's point of view can be assessed and the uncertainties considered quantitatively in the event control strategies are to be based on the models.

104.4 On-Site Computation and Data Display

Objective

The objective of this subtask is to provide computational and display capability at the RAPS central facility to evaluate selected portions of network data for applicability to simplified models.

Purpose and Scope

Although the principal computations for data processing and analysis will be performed in Durham, a need will exist for the preliminary evaluation of measurements, experimental results, and computational models on site in near real time. One aspect of this evaluation can be met simply by providing for a convenient "quick look" at data. Beyond this, there should be facilities for making computations on the basis of simplified models for selected portions of observational records. In this manner, the quality of results can be monitored and improved with minimal delay, and flaws in sensors, techniques, or logic can be detected and remedied early in the RAPS. The computational and display facility should be incorporated into the computational system required for the balloon-tracking facility.

200 Atmospheric, Chemical, and Biological Processes

Objective

The objective of this series of tasks is to develop an improved understanding of the chemical, physical, and biological processes that are entailed in determining the concentration (the dispersal) of pollutants and the modification of air quality.

Purpose and Scope

Purposes of these tasks are to:

- Investigate in a number of parallel programs the various mechanisms entailed in the transport, transformation, and removal of pollutants not now well understood.
- Develop techniques of describing (or better describing) such mechanisms so that they can be accounted for in existing models or models to be developed to accommodate them.
- Identify conditions or processes that are significant in formulating control and abatement strategies to provide air quality amelioration.

The approach follows three major lines. The first approach is the acquisition of a better quantitative knowledge of processes already recognized as significant but that have not been adequately described for modeling purposes. The second approach is development of a better understanding of the significance of various processes in terms of adequately modeling complex air quality factors. The third approach is investigation of pollutants and pollutant processes that are not yet considered in control strategies, and assessment of their importance so that appropriate strategies can be formulated.

201 Gaseous Chemical Processes

Objective

The objective of this task is to develop an improved understanding of the gaseous chemical processes that are important in determining the concentrations of air pollutants and in the design and specifications of simulation models dealing with the transport of gaseous pollutants.

Purpose and Scope

Purposes of these tasks are to:

- Investigate through a number of discrete but interrelated projects various mechanisms that are important in the transformation and scavenging of gaseous air pollutants.
- Carry out specific sampling programs designed to better describe the concentration field of various important pollutants that are not covered by the regular monitoring system.
- Develop special measurement techniques and automatic instrumentation that can be used to describe in more detail the atmospheric concentration fields of specific air pollutants.
- Relate the processes and conditions observed in the field program to existing or potential simulation models and to abatement strategies.

201.1 Hydrocarbon Analyses and Monitoring

Objective

The objective of this subtask is to determine hydrocarbon reaction products in the atmosphere over the regional study area.

Purpose and Scope

This project area will carry out research and monitoring studies directed toward determining the nature of gaseous hydrocarbon materials and reaction products in the atmosphere. The materials that will be covered in these studies include both primary emissions of hydrocarbon compounds and reaction products that are specifically considered in modeling studies. The program will also be concerned with the primary and reaction constituents and the various scavenging processes that serve to transform them in the atmosphere or to remove them from the atmospheric system. Variations in reaction rates from nonhomogeneous mixing in the lower surface layer because of surface roughness and certain meteorological conditions (gustiness) should be assessed as well.

201.2 Development of Hydrocarbon Classifier Instrumentation

Objective

The objective of this subtask is to develop instrumentation to monitor classes of hydrocarbons in the atmosphere.

Purpose and Scope

Atmospheric reaction studies and modeling programs generally treat hydrocarbon compounds as classes rather than specific compounds. Analytical procedures are available for monitoring hydrocarbons as classes rather than specific compounds, and these techniques are amenable to automatic instrumentation and, therefore, to the development of field instrumentation. However, instruments are not now available to carry out these tasks. The regional study program should support the development of instrumentation that will monitor hydrocarbon classes, because these data are necessary for the development and verification of reactive chemical models and appear to be well within the present state of the art.

Note: At the conclusion of the development phase, a decision will be made whether to procure instruments for routine use in the facility. Provided satisfactory automatic instruments can be procured, it is anticipated that they will be installed and operated at 17 Class A stations and 10 selected Class B stations.

201.3 Total Aldehydes-Formaldehyde Monitoring Program

Objective

The objective of this subtask is to determine daily variation in concentrations of total aldehyde and formaldehyde in the region study area.

Purpose and Scope

Aldehydes and in particular formaldehyde are products of atmospheric photochemical reactions and data on concentration patterns are necessary for the development and verification of photochemical reactive simulation models.

201.4 Determination of Peroxyacetyl Nitrate

Objective

The objective of this subtask is to carry out a measurement program of atmospheric concentrations of peroxyacetyl nitrate (PAN) in connection with studies of photochemical reactions in the regional study area.

Purpose and Scope

PAN is an important end product of photochemical smog reactions and as such is a major subject of photochemical modeling processes. Thus, data on PAN should be obtained for use in the simulation modeling program.

201.5 Ammonia Monitoring Program

Objective

The objective of this program is to determine ammonia concentration trends as air parcels passes through urban areas.

Purpose and Scope

Ammonia plays an important role in scavenging SO_2 and NO_2 from the atmosphere through the formation of ammonium salts. Thus, it is also a factor in aerosol formation processes. Both the gaseous scavenging and aerosol formation processes will be included in comprehensive modeling of pollutant chemical processes.

201.6 CO, SO_2, and NO_2 Mass Flux Measurements

Objective

The objectives of this subtask are to:

- Provide, using aircraft and ground-level measurements, transects through selected vertical planes downwind of St. Louis to permit calculation of CO, SO_2, and NO_2 mass flux.

- Provide a measure of integrated sunlight intensity from both aircraft and ground level in the spectral region responsible for photolysis of nitrogen dioxide.

Purpose and Scope

The long distance transport of pollutants can be considered on the basis of the mass flux across vertical planes normal to the wind. Net loss of total pollutant mass as a function of time and other conditions will indicate the magnitude of removal processes and are needed as input to modeling procedures. Processes that are important can be shown by measurements that attempt to obtain a total mass balance regardless of the atmospheric state, e.g., sulfur could be present either as SO_2 or as sulfate particles. Unaccounted losses probably would be attributed to absorption by soils and other surfaces.

The photolysis of nitrogen dioxide is closely related to the intensity of light in the 2900 to 4000 Å region. Because of selective absorption, the proportion of light in the photochemically active region may vary somewhat independently of the total sunlight intensity. Therefore, light intensity measurements should be made in this spectral region. Existing simulation models do not require these data; however, if such data were available, they could result in improved model simulation.

201.7 Origin of Atmospheric CO

Objective

The objective of this subtask is to determine atmospheric CO by C^{12}/C^{13} isotope ratio measurements.

Purpose and Scope

The atmospheric and environmental cycle of CO is not well understood. Current data indicate significant biological involvement in scavenging processes and possibly as a source of natural CO. Studies that can lead to the solution of the CO cycle will contribute to an understanding of the long term global impact of air pollution.

201.8 Atmospheric Odor Identification

Objective

The objective of this subtask is to carry out chemical identification studies to characterize odors observed in urban and rural areas.

Purpose and Scope

The information obtained in the sampling studies and other identification procedures will be used in conjunction with source inventory information to define the extent of specific odors and sources and their impact on the local community. This information in time will be used to provide basic data for the development of control strategies dealing with urban odor problems.

202 Atmospheric Aerosol Processes

Objective

The objective of this area of the program is to develop improved understanding of, and expanded data on, the nature of atmospheric urban aerosols and the processes that are important in determining (1) the chemistry and concentrations of these materials and (2) the application of this information in the design and specifications of simulation models dealing with the transport and transformation of atmospheric aerosols.

Purpose and Scope

The purposes of this task are to:

- Investigate through a number of individual but interrelated projects the various mechanisms that are important in the formation, transformation, and scavenging of atmospheric aerosol particles.

- Carry out specific sampling programs designed to better describe both the chemistry and concentration fields of various atmospheric aerosols and to relate these measurements to the routine measurements obtained by the regular monitoring system.

- Conduct a coordinated program of gaseous and particulate sampling experiments with the goal of describing the specific mechanisms by which atmospheric aerosol particles are formed from gaseous contaminants.

- Develop special measurement and instrumentation techniques that through automatic operation can be used to describe in more detail the atmospheric concentration fields of specific particulate materials.

- Relate the processes and conditions relative to the formation and transport of atmospheric aerosols to existing and future simulation models and to abatement strategies.

The individual projects within this program area fall into three general categories: (1) special monitoring programs for specific aerosol materials, including size distribution studies, aerosol chemistry, and studies of the interaction of aerosols and gaseous contaminants; (2) development of instrumentation and analytical techniques to improve the effectiveness of aerosol monitoring programs and to provide the data necessary for simulation modeling applications; and (3) special transport and scavenging research studies aimed specifically at delineating atmospheric processes that serve to determine the characteristics of atmospheric aerosols in urban and rural areas.

For the most part, research in atmospheric aerosols is hampered by the fact that relatively little automatic instrumentation can be used to obtain detailed data on the chemical constituents that make up the atmospheric mass. As a result, most of the programs are based on a gathering of specific field samples followed by a period of laboratory analysis to determine the nature of the collected samples. This means that the samples generally have poor time resolution because of the length of time necessary to collect enough material for adequate analysis, and because the number of samples that can be collected practically is limited because of lack of available manpower and laboratory facilities. If suggested instrumentation development is successful, some of these problems may be ameliorated; however, since the suggested instrumentation is not new but has been recognized for a number of years, a higher degree of hope cannot be expressed for the successful resolution of this instrumentation design problem.

202.1 Determination of Total Nitrate in Aerosol Samples

Objective

The objective of this subtask is to determine the concentration of nitrate in aerosols collected on high-volume sampler samples.

Purpose and Scope

One of the scavenging processes for NO_2 is considered to be the formation of NO_3^- salts. If a satisfactory modeling program is to be undertaken for the dispersion of NO_2, then this conversion to an aerosol must be considered. The process must be considered as being relative to other ambient conditions such as sunlight and humidity.

202.2 Determination of Total Sulfate in Aerosol Samples

Objective

The objective of this subtask is to determine the concentration of sulfate in aerosols collected on high-volume sampler samples.

Purpose and Scope

One of the scavenging processes for SO_2 is considered to be the formation of $SO_4^=$ salts. If a satisfactory modeling program is to be undertaken for the dispersion of SO_2, then this conversion to an aerosol must be considered. The process must be considered as being relative to other ambient conditions such as sunlight and humidity.

202.3 Determination of Aerosol Size-Distribution

Objective

The objective of this subtask is to determine aerosol size distribution under a variety of atmospheric conditions.

Purpose and Scope

The research will obtain detailed data on the size distributions of the total atmospheric particulate material and on specific chemical fractions. The data should apply to a variety of ambient conditions and coordinated with other data gathering programs so that the mechanisms governing the areawide distribution of urban and rural aerosols can be obtained. These mechanisms will then lead to model development of aerosol formation and dispersion.

202.4 The NO_2 NaCl Reaction in Aerosol

Objective

The objective of this subtask is to make a preliminary estimate of the importance of the NO_2 - NaCl reaction in aerosols and recommend the need for more extensive programs.

Purpose and Scope

One process proposed as a scavenging process for NO_2 is its reaction with NaCl in droplets. The result is the release of chlorine. This program is exploratory to determine whether chloride is prevalent enough in the RAPS area to make this mechanism worthy of further consideration.

202.5 Isotope Ratios of Sulfate Aerosols

Objective

The objective of this subtask is to develop information on the possible sources of atmospheric sulfate aerosols on the basis of sulfur isotope ratio analyses.

Purpose and Scope

Sulfate aerosols are an important component of urban and natural atmospheric aerosol systems and can result from several different sources, namely, the oxidation of pollutant generated SO_2, the oxidation of H_2S from natural sources, and from residual sulfate contained in marine aerosols. These several sources have different ratios between the two sulfur isotopes S^{34} and S^{32}. On the basis of the isotope ratios found in atmospheric aerosols, inferences can be drawn concerning the sources of these aerosol materials. Thus, these studies will be directed to provide increased information on the probable sources of atmospheric aerosol particles.

202.6 Organic Compounds in Particulate Material

Objective

The objective of this subtask is to develop data on the specific organic compounds that are present in urban and rural particulate samples.

Purpose and Scope

Organic compounds of a wide variety make up a major share of the atmospheric particulate material; however, relatively little is known about the specific compounds that constitute this total mass of organic material.

To better understand the formation mechanisms and the importance of various sources of urban and rural haze, it is important to know what specific materials are present in the atmospheric aerosols. After the nature of the various components are known, it should be possible to assess the importance of various urban and rural sources as contributors to pollutant haze concentrations and to include relevant mechanisms in simulation models and control strategies.

202.7 Experimental Measurements of Deposition Velocity

Objective

The objective of this subtask is to determine through direct experimental measurements the apparent deposition velocity applicable to atmospheric aerosol particles of different chemical constituents.

Purpose and Scope

Atmospheric particles, especially those in the small-size ranges, and particularly less than one micron in diameter, deposit on surfaces at a rate that is not always predictable on the basis of the physical size of the particle and the accepted Stokes' settling velocity. Since ground deposition is an important mechanism by which particles are removed from the atmosphere, it will be extremely useful to measure the apparent deposition velocity that is applicable to various atmospheric particles and to different types of ground surface. Once deposition velocities are determined that are applicable to atmospheric aerosol particles, this term can be introduced into simulation modeling formulas and used to estimate the dispersion of aerosol materials and subsequent concentration patterns.

203 Other Pollutant Related Atmospheric Processes

Objective

The objective of this task is to develop an improved understanding of a wide range of atmospheric processes that are important in understanding the transport, transformation, and final removal processes of atmospheric pollutants.

Purpose and Scope

The research effort will investigate through a number of individual research projects various atmospheric processes and mechanisms that are related to the understanding of pollutant distribution over an urban and rural area. These atmospheric processes cover a range of applicable conditions and have not been readily classifiable into other areas of this research prospectus.

The individual research projects carried out within this section, while dealing with specific and identifiable objectives, will generally relate in some detail to one or more of the other research projects described in other parts of this Prospectus.

203.1 Radiation Balance Studies

Objective

The objective of this subtask is to assess the influence of urban areas on the radiation balance of the lower atmosphere.

Purpose and Scope

The purpose of the subtask is to:

- Document differences in urban and rural radiation budgets for the surface and boundary layer along with differences in pollutant concentrations, temperature, water vapor, and cloudiness.
- Provide radiation data required for the formulation of direct input into applied mathematical models of the boundary layer.
- Evaluate, modify and verify pertinent radiative components currently used in mathematical models.

203.2 Visibility Reduction in Urban and Rural Areas

Objective

The objective of this subtask is to develop quantitative data on urban visibility restrictions and to determine from relationships with aerosol particle studies the mechanisms and processes responsible for observed reduced visibility conditions.

Purpose and Scope

Concentrations of urban haze that result in visible pollutant concentrations are an important aspect of urban air pollution problems. Although these are directly relatable to atmospheric aerosol concentrations and perhaps to the concentrations of visible gaseous pollutants such as NO_2, it is important to analyze this concentration data in terms of observed or expected visibility reduction. Only by relating the technical findings to the conditions reviewed by the general population of an area can the technical program be put on a practical basis. After pollutant concentration data and other factors are related to the urban visibility restriction problem, simulation modeling techniques that tackle specifically the problem of visibility restriction can be developed and subsequently verified.

203.3 Transport of Atmospheric Odors

Objective

The objective of this subtask is to determine the specific transport phenomena that may relate directly to the transport of atmospheric odors and in particular to the long distance transport of identifiable odors.

Purpose and Scope

Odor identification apparently does not require that the odor concentration exists for a long period of time to be detected. Thus, atmospheric studies and transport mechanisms, whereby even relatively small volumes of gas can remain at high concentrations even though an average integrated concentration is extremely low, can result in the long distance transport of identifiable odors. If it were possible to characterize the conditions under which long distance odor transport was significant, specific modeling techniques could be developed for odor detection and odorous sources. These subsequently could be used in the design of control strategies and air quality criteria related directly to odor sources.

203.4 Trace Metals and Toxic Trace Materials

Objective

The objective of this subtask is to determine in detail the distribution and atmospheric concentrations of trace metals and other toxic trace materials throughout the regional study area.

Purpose and Scope

The purposes of this research are to develop information on the extent to which toxic trace materials such as beryllium, mercury, cadmium, and asbestos are distributed throughout the Regional Study area and, in particular, to determine the relationship of identifiable sources to atmospheric concentration patterns and to deposition rates onto surfaces, vegetation, and bodies of water. Lead aerosols from the combustion of vehicular fuels should also be considered in this program, although these materials are so widespread in both the urban and rural areas that it is questionable whether lead aerosols can be considered as a trace material in the same manner as cadmium or beryllium. Once the distribution of these materials is characterized, urban area exposures can be determined and control strategies designed.

203.5 Agricultural Chemical Distribution

Objective

The objective of this subtask is to determine the distribution and concentration patterns of agricultural chemicals throughout the Regional Study areas, including both agricultural and urban areas.

Purpose and Scope

A wide range of agricultural chemicals, including pesticides, herbicides, and fertilizers, are applied as sprays from aircraft over large areas of agricultural land, and little is known about the transport and the concentrations of these materials beyond their area of application. However, considerable concern is shown about the adverse effect that may result from widespread distribution of agricultural chemicals in both rural and urban areas. Once data are obtained on the areawide distribution of agricultural chemicals and related to the nature of their application, control strategies can be devised to protect the community from indiscriminate use of agricultural chemicals.

203.6 Natural Sources of Air Polluted Compounds

Objective

The objective of this subtask is to determine the types and magnitudes of sources within the natural environment of chemical compounds that are also emitted as air pollutants from urban and industrial sources.

Purpose and Scope

Practically all the materials that are classed as air pollutants because of their emission from urban and industrial pollutants sources also have sources within the natural environment; although these are almost always incapable of producing significant concentrations of material, their total contribution on a global atmospheric basis to the atmospheres chemistry is usually calculated as being very significant. Within an urban area, the existence of natural sources of contaminant materials results in a very low background level being observed. Thus, to complete the description of the areawide distribution of urban pollutants, it is highly desirable to be able to include recognition of the presence of this natural low level background concentration. In addition, careful analysis of background pollutant emissions may also provide clues as to the most effective scavenging mechanisms that serve to remove these materials from the atmosphere, including pollutant generated materials.

204 Atmospheric Scavenging by Precipitation

Objective

The objective of this task is to develop an improved understanding of the nature of precipitation scavenging of atmospheric pollutants and to relate these processes to other environmental factors such as water quality.

Purpose and Scope

Purposes of this task are to:

- Investigate through sampling and analytical projects the extent and processes of precipitation scavenging of atmospheric pollutants.
- Relate the observed precipitation scavenging processes to pollutant emissions and downwind pollutant concentration patterns and to develop models by which the impact of precipitation scavenging can be included in simulation modeling of the transport and dispersion of atmospheric pollutants.
- Develop special instrumentation where necessary to sample and analyze precipitation for scavenged air pollutants.

The individual projects within this program area include instrument development and sampling programs directed toward the measurement of precipitation chemistry. After analytical results are available, the data will be related to air quality concentration patterns and to meteorological conditions to develop a better understanding of the nature of the precipitation scavenging process.

204.1 Instrument Development for pH and Chemical Sampling

Objective

The objective of this subtask is to develop the instrumentation necessary to include pH sampling and chemical sampling of rainfall in the regular monitoring stations within the Regional Study network.

Purpose and Scope

It is desirable to obtain data on precipitation pH and chemical content on a more or less routine basis at the regularly reporting Regional Study network stations. Suitable standard equipment is not available for these measurements, and special designs suitable for the Regional Study network will have to be developed.

204.2 Rainfall pH Measurements

Objective

The objective of these measurements is to determine the spatial variation of the acidity of rainfall in the St. Louis regional area.

Purpose and Scope

The pH of rainfall is often used to indicate the effect of atmospheric pollutants on rainfall chemistry. This conclusion is a result of the fact that many atmospheric contaminants, especially SO_2 and NO_2, lead to the formation of ascetic particulate material which is readily incorporated in precipitation and thus can affect the pH of the precipitation. Thus, an areawide distribution pattern of rainfall pH can be used to indicate the impact of urban area pollutant sources on precipitation. These data will provide another indication of the magnitude of the transport of pollutants from the urban area and their possible impact on the surrounding environment.

204.3 Measurement of Rainfall Chemistry

Objective

The objective of these measurements is to determine the chemical constituents contained in precipitation samples in the Regional Study area.

Purpose and Scope

Details on the precipitation scavenging process for atmospheric pollutants can be obtained by making chemical analyses for various materials on samples of collected precipitation. With these data and corresponding air quality and meteorological information, important factors describing the precipitation scavenging process for specific pollutants can be obtained. Once the precipitation scavenging process has been described, it can be incorporated in simulation models and lead to an improvement in the simulation modeling process for the distribution and transport of atmospheric pollutants.

205 Air Pollutant Scavenging by the Biosphere

Objective

The objective of this task is to develop an improved understanding of the relationship of the biosphere to air pollutant dispersion and transport and in particular the effect that the biosphere has on the scavenging of air pollutants from the atmosphere.

Purpose and Scope

Atmospheric transport and dispersion processes serve to bring air pollutants in contact with large amounts of biological material, and it is known that plants are effective absorbers of trace chemical materials from the atmosphere. This program will conduct projects designed to provide quantitative estimates of the scavenging mechanisms that are effective within the biosphere in removing air pollutants from the atmosphere.

The research effort also will relate the scavenging processes observed in the field to existing or potential simulation models and to an evaluation of the interaction between air pollution and the biosphere.

The individual projects within this program area will include chemical analyses of vegetation material to determine the extent to which absorbed pollutants are reflected in changed chemical composition and special monitoring studies to determine the extent to which vegetation absorption can be observed to be reflected in actual changes or anomalous concentration patterns in the atmosphere. Once quantitative values are obtained for the scavenging of vegetation materials for air pollutants, the relationships can be interpreted in terms of simulation model parameters and used as a basis for further research studies.

205.1 Chemical Content of Vegetation

Objective

The objective of this analysis is to determine the concentration of various pollutant chemicals in plant materials growing in areas exposed to a variety of pollutants and pollutant concentrations.

Purpose and Scope

When vegetation is exposed to contaminated atmospheres, it is common for the pollutant material to be detected in the vegetation itself, and the content of vegetation often is different for different species or types of material even though the exposures are essentially the same. Pollutants for which changes in vegetation concentration might be expected and that could provide additional measurements on the distribution of these contaminants include lead aerosols, fluorides, and perhaps some of the heavy trace metals. The more soluble materials and materials involved directly in plant metabolism such as sulfur and sulfate are unlikely candidates for plant surveys because of the relatively high concentrations already present in the vegetation and the likelihood that atmospheric concentrations would be completely masked by this background concentration.

205.2 Atmospheric Pollutant Concentrations Related to Vegetation Absorption

Objective

The objective of this subtask is to determine on the basis of atmospheric sampling the extent to which travel across various types of vegetated surfaces causes anomalous reductions in the concentration of air pollutants.

Purpose and Scope

During its life cycle, vegetation has a large intake of atmospheric air and this forms a very large part of the plant life cycle. During the intake of atmospheric air, pollutants are also brought into the plant where they are retained; in addition, plant surfaces are exposed to the flow of air, and as such they provide areas where pollutants may be deposited or absorbed even though not being brought directly into the plant tissues. These are loss mechanisms that in some cases have been shown to be significant and to cause measurable changes in the ground level concentrations of specific air pollutants. This research project will attempt to quantify these absorption or removal mechanisms.

206 Atmospheric Processes

Objective

The objective of this task is to provide an additional understanding and description of physical atmospheric processes not already accounted for in generalized boundary layer meteorological modeling.

Purpose and Scope

The research for this task will extend the capability of general boundary layer theory to include anomalous physical factors of significance in the dispersal of pollutants. These factors include smaller scale phenomena, such as the effects of special emission conditions (i.e., at the stack) on the behavior of effluent plumes, or the local variation of surface roughness (i.e., different terrain or land use conditions) on air flow--or larger scale phenomena, such as the influence of extraregional features or synoptic scale circulations on the air flow characteristics within the Region.

Inputs will be obtained from other ongoing research programs or from special studies conducted within the RAPS program. Consideration will be given to conducting physical modeling studies in the laboratory where appropriate.

206.1 Source Factors in Pollutant Dispersal

Objective

The objective of this subtask is to attain a better understanding of the nature and significance of source factors and processes and their relationship to the dispersion of effluents.

Purpose and Scope

The purpose of this research area is to achieve an improved capability to describe through simulation modeling the interactions and ramifications of various source-related factors, e.g., source height, efflux velocity and buoyancy, aerodynamic characteristics of stack and local environment, moisture, and plume opacity and composition. Additionally, the program should seek to examine the integrated effect of anthropogenic processes on diffusion characteristics and the depth of the atmospheric mixing layer.

206.2 Terrain and Surface Roughness Effects

Objective

The objective of this subtask is to attain an improved understanding of the effects of topographical surface characteristics on the transport and dispersion of effluvia.

Purpose and Scope

The purpose of this subtask is to achieve the capability to describe and predict the effects of surface features on atmospheric dispersion processes in general. In particular, the program will seek to understand and model the effects on the eddy diffusivity, the near-surface vertical veering of the wind, the effective horizontal transport wind speed, the mean and fluctuating nature of the vertical component of the wind, horizontal veering (curvature) of the wind field, the depth and structure of the mixing layer, and the temporal and lateral variability of these factors.

206.3 Extra Regional and Synoptic Scale Circulations

Objective

The objective of this subtask is to develop techniques of accounting for extraregional factors and synoptic scale effects on the meteorology and air flow patterns of the regional area.

Purpose and Scope

The research in this program will ascertain to what extent and in what manner macroscale factors contribute to regional meteorology and climatology in terms of the air flow patterns or the modification of air masses as a function of the upstream terrain or orography.

Although this subtask is primarily concerned with physical factors such as wind trajectories, stability, and moisture content, consideration also will be given to the effects of the upwind history on the condition of the air entering the regional area--in terms of both natural factors such as dust loading and man-made pollution content.

300 Human, Social, and Economic Factors

Objective

The objective of the tasks in this series is to develop a better understanding of factors of significance to the design of improved control strategies in the urban/rural complex, including health and economic effects and the role of land use and community planning.

Purpose and Scope

Purposes of this research program are to take advantage of the unique facility the RAPS organization provides to collect data on human, social, and economic factors in an economical and well focused manner to complement the purely physical aspects of RAPS, for the subsequent formulation of improved control strategies. The data will provide the additional basis that will be needed to apply the lessons learned in the RAPS in improved control strategies that are effective and acceptable in terms of priorities, costs of implementation, and value of results.

The resources of field teams, data analysts, and data processing facilities will be made available to collect human, social, and economic data identified as significant to the purpose noted. Data will be collected either by adding elements to other data collection surveys or by initiating special surveys. Steps will be taken to ensure that the data are compatible with the physical data collected so that interpretation and evaluation of the data on effects will be facilitated.

301 Human and Social Factors

Objective

The objective of this task is to provide a data base of relevant information on human and social factors that can be used in RAPS and in developing methodologies for using such data in other areas.

Purpose and Scope

Purposes of the task are to:

- Take advantage of the data handling and analysis capabilities of a RAPS organization to gather data on epidemiology, mortality, and the like.
- Determine population concentrations and land use characteristics.
- Provide information on the utilization of the labor force and its skills and mobility.

301.1 Data on Epidemiology and Health Effects

Objective

The objective of the subtask is to develop quantitative data describing the characteristics of the population in the St. Louis area in terms of epidemiology, health, and demographic factors that are known or believed to be interrelated with air quality.

Purpose and Scope

The RAPS area will be defined in unprecedented detail in terms of meteorology, air quality, and emission sources. These, and perhaps other data, will be used intensively for the scientific purposes of model verification and related goals. Many of these data can be utilized in the analysis of other air pollution problems, such as the probable effects on the health of the population. Accordingly, this task is designed to acquire detailed data covering the status of the health and related factors of the resident population and to couple these data with the appropriate aerometric data using the St. Louis data handling facilities. These data generally would be designed to support ongoing research efforts elsewhere in EPA not currently considered as part of RAPS.

301.2 Data on Population and Land Use Characteristics

Objective

The objective of this subtask is to develop quantitative data describing the characteristics of the population distribution and land use patterns in the St. Louis area.

Purpose and Scope

The RAPS area will be defined in unprecedented detail in terms of meteorology, air quality, and emission sources. These, and perhaps other data, will be used intensively for the scientific purposes of model verification and related goals. Many of these data can be used in the analysis of other air pollution problems, such as the correlation of land use patterns and population distributions with air quality. This task is designed to acquire detailed data covering these characteristics and related factors and to combine these data with the appropriate aerometric data using the St. Louis data handling facilities. These data generally will be designed to support ongoing research efforts elsewhere in EPA not currently considered as part of RAPS.

301.3 Data on Labor Force Utilization

Objective

The objective of this subtask is to develop quantitative data describing the characteristics of the labor force in the St. Louis area in terms of skills, mobility, and patterns of utilization.

Purpose and Scope

The development of profiles of labor force utilization in the St. Louis area will provide the basis of assessing the impact of alternative control and abatement strategies on employment. The mix of gained and lost employment opportunities can be developed for selected strategies, with the results formulated in both economic and social terms. These data generally will be designed to support ongoing research efforts elsewhere in EPA not currently considered as part of RAPS.

302 Economic Factors

Objective

The objective of this economic research is to provide a data base of relevant information on economic factors that can be used in RAPS and in developing methodologies for using such data in other areas.

Purpose and Scope

The purpose of the task is to take advantage of the data handling and analysis capabilities of the RAPS organization to gather data on the various costs of air pollution to the industrial and general population (e.g., depression of property values, damage to property, loss of productivity due to sickness) and also the cost of air pollution control strategies, in terms of both plant modification and increased costs of production.

302.1 Costs of Inferior Air Quality to Industrial and General Population

Objective

The objective of this cost study is to develop quantitative data to provide measures of the cost of inferior air quality.

Purpose and Scope

The costs of low quality air are revealed in a variety of forms, including medical costs, job absenteeism, reduced property values, increased maintenance, and more rapid deterioration of materials. The RAPS data bank on regional air quality will provide a heretofore unavailable opportunity to carry out detailed analyses of the costs of inferior air quality. This task is designed to acquire detailed data covering these and other pertinent costs and related factors of the resident population and industry and to combine these data with the appropriate aerometric data, using the St. Louis data handling facilities. These data generally will be designed to support ongoing research efforts elsewhere in EPA not currently considered as part of RAPS.

302.2 Costs of Control Strategies

Objective

The objective of this subtask is to assemble information to support analyses of the costs of alternative control strategies.

Purpose and Scope

An immense number and variety of alternative control strategies have been identified, including installation of various types of control equipment, changes in fuel usage, plant shutdown, traffic controls, and land use restrictions on source densities. Each has a cost associated with its implementation, and many of these costs are not known to a satisfactory level of accuracy. Accordingly, this task covers the development of the necessary data to support further studies of the cost of control strategies. The collection of these data should be coordinated with other collection tasks, especially with 102--Source Inventories. These data generally will be designed to support ongoing research efforts elsewhere in EPA that are not currently considered as part of RAPS.

302.3 Data Collection Surveys of Specific Effects

Objective

The objective of this subtask is to assemble information to support analyses of specific effects of pollutants on selected receptors.

Purpose and Scope

The effects of pollutants on receptors naturally vary widely with the nature of each. Typical effects may include corrosion of metals, hardening and cracking of electrical insulation, and damage to growing plants. The nature of the damage will be quantified and where possible combined with the RAPS air quality data. These data generally will be designed to support ongoing research efforts elsewhere in EPA not currently considered as part of RAPS.

400 Transfer of RAPS Technology for Control Agency Applications and the Formulation of Control Strategies

Objective

The objective of this series of tasks and supplies is to develop improved technology that can be applied in local and regional control agency operations, including techniques for emission inventories, air quality and meteorological measurement, data handling and analysis, and the objective assessment of control strategy effectiveness.

Purpose and Scope

The purpose of this research area is to ensure that the knowledge and technology developed in the RAPS is transferred widely to the air pollution control community at large as early and effectively as possible. This requires passing on improvements and innovations in the techniques of measurement, data handling, utilization in a direct form. It also includes the conversion and distillation of the knowledge and technology developed in RAPS in the specific test region to a generalized form, so that it can be applied in other regions and for other problems with minimum difficulty. Above all, however, this task provides the basis for the development of improved control strategies, by national, state and local agencies.

The approach follows four major lines. The first approach is development and description of techniques and criteria by which the basic air pollution factors can be assessed and monitored on an operational (as distinct from research) basis. Particular attention will be given to identifying and developing new techniques of monitoring atmospheric and air quality conditions on extended scales appropriate to regional and subregional control strategies so that the costs may be minimized. The use of aircraft and remote probing techniques either from such aircraft, or from the surface are especially suited to this purpose and every attempt should be made to advance their applicability.

The second approach is the provision of tested, effective simulation models, suitable for operational use on a generalized basis (that can be readily modified and adapted),for other areas and conditions. The third approach is development of a methodology for assessing the validity (in terms of confidence, accuracy, and precision) of the preferred models, for varying degrees of input data quality. The fourth approach is to provide methodologies for determining and assessing other factors such as health effects and economic costs and benefits relevant to the formulation of improved control strategies.

First priority will be to provide data applicable to current data resources, deficient though these may be, with emphasis on the optimum methods of providing more complete data for input to models and for air quality monitoring and model verification purposes. The needs of the states and local authorities (and EPA) to improve and extend Implementation Plans will be treated first, with concurrent although subordinate attention to Environmental Impact Statement requirements. First milestone will be the achievement of interim capabilities for these purposes. Thereafter, a more complete and detailed facility will be developed, covering all aspects of control strategies formulation then current and capable of extension to other pollutants and the like as the need arises.

401 Source Inventory Procedures

Objective

The objective of the task is to provide guidelines for the development and maintenance of source inventories as derived from the experience in the Regional Study.

Purpose and Scope

The source inventory for the Regional Study is likely to be the most comprehensive such inventory yet developed. The methods by which the source data are acquired and updated, and the manner in which the inventory is organized, stored, and retrieved should prove to be of considerable value to others faced with the task of preparing sources inventories. Accordingly, this component of the Research Plan includes efforts required to prepare guidelines and reports for the benefit of others, covering the techniques developed in the Regional Study for the management and use of source inventories.

401.1 Techniques for Making Source Inventories

Objectives

The objective of this subtask is to provide guidelines for the preparation of source inventories.

Purpose and Scope

The development of the source inventory for the Regional Study is likely to reveal many near optimal techniques by which all but the most unique sources should be inventoried. For example, requirements for field measurements of the various types of sources versus the more economical on-site inspection and similar procedures should be readily determined. The most appropriate classification scheme for the many types of sources should also be apparent. These and other findings should be provided to all organizations confronted with responsibilities for the development of emission inventories whether for research control, strategy design, or enforcement.

401.2 Techniques for Inventory Storage and Retrievel

Objective

The objective of this subtask is to provide guidelines for the development of techniques for source inventory storage and retrieval.

Purpose and Scope

The magnitude of the source inventory of the Regional Study undoubtedly will require the use of electronic data processing equipment. Because of the many anticipated uses of the inventory during the work, the storage and retrieval techniques used to treat the inventory should be highly flexible and have the capability to provide the required data to each user in almost any form requested. These techniques should have ready application to emission inventories maintained by other research groups, control agencies, and the like. Therefore, these groups should be provided with both the concepts and the computer programs developed in the Regional Study for application elsewhere.

401.3 Techniques for Updating the Source Inventory

Objective

The objective of this subtask is to provide guidelines for updating source inventories.

Purpose and Scope

Tasks 102.2 and 102.3 provide for the development of inventories for stationary and mobile sources for the Regional Study and the maintenance of these inventories throughout the study. This maintenance program should provide measures of the rate of change of emission levels over the five-year period for all important classes of sources. Moreover, the causes of the change should be available. While perhaps the St. Louis area is not precisely typical of other urban areas, valuable clues of inventory changes based on the St. Louis experience should be provided to others as guidance for updating their inventories.

401.4 Relating Source Inventory to Control Strategy

Objective

The objective of this subtask is to provide guidelines for the development and maintenance of source inventories to support control strategies.

Purpose and Scope

The design and implementation of control strategies require some form of source inventory. A difficult fact to ascertain in this regard is the required size and level of detail of the inventory. The source inventory of the Regional Study perhaps will be the most complete and detailed inventory yet developed. Appreciable costs are associated with inventory development, but these costs are justified for the comprehensive Regional Study. On the other hand, in view of the number of areas in the nation for which control strategies may be designed, the cost and time to develop an emission inventory equivalent to St. Louis may be unacceptable and possibly unnecessary.

To develop guidelines for inventory development to support control strategies, the comprehensive Regional Study inventory should provide the opportunity to develop alternative control strategies based on use of the inventory at successive levels of detail. The level of detail will vary as a function of the number of sources included and the time interval over which emission levels can be characterized, such as annually, daily, or hourly. Comparison of the potential success of each control strategy should reveal the required level of detail required in a source inventory.

402 Atmospheric Monitoring

Objective

The objective of this task is to improve the technology and technique of monitoring atmospheric conditions, particularly on extended scales, so that control strategies can be better implemented.

Purpose and Scope

The purpose of this task is to make available for use in all types of air pollution control operations as early as possible, the lessons learned and the techniques developed in the RAPS.

Basic principles for designing measurement networks for control agency operation and criteria for the siting of monitoring stations and instrument exposure, will be developed on the basis of experience with the RAPS data collection network.

Criteria for the organization and maintenance of extended networks of measuring instruments, with special reference to calibration and standardization, will be established. This research effort also will develop a methodology by which newly acquired data, using new techniques, can be related to older data so that the value of the latter is fully realized, even if strict continuity is not maintained.

Remote probing systems will be tested and evaluated, in comparative trials as candidate systems become available. Such tests will be conducted in conjunction with both the standard RAPS data collection system and special programs that provide especially detailed knowledge of meteorological conditions or the concentration of pollutants.

In particular, use will be made of helicopter soundings and other aircraft acquired data. The application of remote probing techniques from aircraft, as well as of aircraft in situ measuring techniques, is an additional and important study subject.

402.1 Basic Network Principles

Objective

The objective of this subtask is to develop and define the basic principles applicable to the design and implementation of a monitoring twork that will provide representative data concerning the meteorology nd air quality of a region. Data manipulation required to achieve the information goals of a monitoring network also will be defined.

Purpose and Scope

The purpose of the research effort is to make available the experience gained in the RAPS in the design, implementation, and operation of an extensive monitoring network and to develop principles and criteria that can be applied to the establishment or modification of a monitoring network by control agencies.

The desirability of three dimensional measurements in the surface layer will be defined on the basis of measurement significance with respect to cost.

Data acquisition techniques, data analysis, and the data format for ease of archiving and retrieval will be characterized in terms of versatility, cost, and applicability to the more limited networks of local control agencies.

The principles and guidelines of monitoring network function will be developed with the required flexibility to find application not only with today's information goals but also with future goals.

402.2 Criteria for Organization and Maintenance of Observational Networks

Objective

The object of this subtask is to develop criteria for the organization and maintenance of observational networks applicable to control agencies whose region of responsibility is extensive and complex in terms of source emissions and location, topography, and land usage.

Purpose and Scope

Criteria for organization, function, support functions, and the mechanics of implementing an extended network will be established on the basis of experience with the RAPS data collection network.

Maintenance, downtime, station calibration, interstation calibration, station accessibility, and instrument shelter parameters will be defined where appropriate in terms of frequency, accuracy, cost, difficulty, and time.

402.3 Station Siting and Instrument Exposure Criteria

Objective

The objective of this subtask is to define criteria for the optimum location of fixed and mobile stations and for the choice, exposure, and operation of meteorological and air quality sensors.

Purpose and Scope

Purposes of this research effort are to:

- Ensure that the measurements obtained at an atmospheric monitoring station are representative of average conditions over a reasonably extensive area and do not reflect the influences of extremely local factors.
- Determine the sampling frequency and period that is required for the various sensors as dictated by the physics of the processes being monitored and the models or studies that will use the data.
- Define objective criteria for the station density that is required for various parts of the region (e.g., urban or rural) and the requirements of special research studies.

402.4 Methodology for Modernization of Monitoring Networks

Objective

The objective of this subtask is to use the RAPS monitoring network with its sophisticated instrumentation as a reference standard against which to compare older design pollutant monitors and their inaccuracies resulting from interferer compounds. Included in the comparison will be new and promising monitors based on new or different measurement techniques and other aspects of network operation such as data acquisition, calibration stability, sample handling.

Purpose and Scope

The purposes of this subtask are to identify older design monitors still in use whose measurement characteristics are such that upgrading with modern instrumentation would be recommended, to identify and define the inaccuracy of monitors resulting from interferer compounds, and to identify new and promising measurement techniques. The data obtained precisely could define the older instrument measurement characteristics and the contribution of interferers. This information can be used to reevaluate data from previous aerometric surveys and to permit the survey data to be upgraded without repeating the measurements.

402.5 New Aircraft Measurement Techniques

Objective

The objective of this subtask is to test and evaluate new aircraft measurement techniques as a means of obtaining meteorology and air quality data over extended areas with three dimensional definition.

Purpose and Scope

Purposes of the subtask are to:

- Make available the experience gained in RAPS in using new and promising airborne measurements.
- Determine the types of airborne measurements that can add significant data basic to the understanding, meteorology, and air quality of a region.
- Evaluate new instruments as to their applicability of airborne operation and correlate their measurments to current instrumentation.
- Define those aircraft measurement techniques most feasible for augmentation of local control agency ground based monitoring networks in terms of expense and adaptability to light aircraft.

402.6 Evaluation of Remote Measuring Techniques

Objective

The objective of this subtask is to test and evaluate remote measuring techniques and to correlate remote probing data with those obtained by ground based and airborne monitors. It will also be determined if the integrated remote measurements can provide measurements representative of the air quality within a region.

Purpose and Scope

Purposes of this research effort are to:

- Make available the experience gained in RAPS in the operation, accuracy, and interpretation of data acquired by remote measuring techniques.
- Evaluate the capability of remote measurement techniques to integrate pollutant concentrations through both vertical and horizontal planes within a region of interest.
- Evaluate the use of remote measuring techniques as to its potential as a policing or reconnaissance tool for local control agencies.
- Evaluate and correlate pollutant concentration measurements over a similar measurement path obtained with remote probing techniques based on different measurement principles.

403 Data Handling

Objective

The objective of the data output task is to develop optimum techniques for acquiring, storing, and retrieving data on an extended scale for use in air pollution control agency operations.

Purpose and Scope

The purposes of the task are to:

- Make availabe the lessons learned and techniques developed in RAPS regarding the handling of all types of data collected and used in an extensive monitoring network and emission inventory.
- Develop and publish Standard formats as used in RAPS that are suitable for general use.
- Develop and publish manuals and computer programs for all major types of data collection and initial processing, suitable for use in air pollution control agency use.
- Provide guidelines on quality control procedures for use in collecting data.

404 Modeling Technology

Objective

The objective of this task is to provide the best available modeling capability for use in operation in air quality management.

Purpose and Scope

The purpose of this task is to extract from the research and experience of RAPS a number of models that have been tried and demonstrated and to show how these can be adapted and used for a range of specific operational requirements in an optimum fashion.

A most important major task is to evaluate the significance of modeling techniques to the formulation of control strategies and their implementation. This entails an assessment of the accuracy and precision of the models output as a function of the degree of completeness of the input. Given that the resources for data collection and monitoring in the general case will be far less complete than those for RAPS, it will be necessary to analyze the way in which limitations of the input can compromise the output of the models. Any shortcomings or uncertainties of the predictions derived from the models must be fully assessed and understood in terms of the control strategies based on them--especially where such strategies have significant economic or social impacts.

In addition to models suited to regional areas in general for the range of significant pollutants, special attention must be paid to providing suitable models for use in formulating or checking Implementation Plans, as well as for use in Environmental Impact Statements, and the requirements of air pollution episode prediction.

For all these purposes it will be necessary to:

(1) Select and publish a series of models relating to appropriate scales or pollutants in a form in which they can be readily applied in an operational role.

(2) Develop and provide a methodology for assessing the sensitivity of such models to practical limitations--such as the quantity or quality of input data, and qualifying topographical features.

(3) Develop and provide a methodology for measuring the accuracy of predictions based on such models, using either existing or specially provided (but limited) additional measurement facilities.

In general the detailed tasks, covered within the 404 series are scheduled well along in the Research Plan at a relatively low level of effort. Accordingly, their ultimate detailed content tends to be somewhat more speculative than tasks presented elsewhere in the Research Plan, so that presentation of their content in detail does not appear warranted at this early time. Broadly, however, the tasks included in the 404 series are as follows:

404.1 The significance of Modeling to the formulation of control strategies and their implementation

404.2 Implementation Plan Applications

404.3 Environmental Impact Statement applications

404.4 Methodology for assessing model validity in Control Agency operations.

405 Other Significant Factors in Control Strategy Formulation

Objective

The objective of the other tasks concerned with strategy formulation is to ensure that all knowledge and experience acquired under the RAPS program is made available for use by the air pollution control community in general and those concerned with formulating improved control strategies in particular.

Purpose

The purpose of these other efforts is to take care that both during the RAPS program and at its conclusion, fullest advantage is taken of other research in progress (both by EPA and other agencies) and also that the products of RAPS not directly connected with its principal objectives nevertheless are made available in appropriate form to potential users. Five principal components are involved.

(1) Liaison and interaction will be required with other research programs, both inside EPA and in other agencies, particularly those being carried out in the St. Louis area, such as METROMEX.

(2) Techniques should be developed to assess and evaluate social and economic factors in control strategy formulation, using as a base the data collected in 301 and 302.

(3) A methodology for assessing operational costs of control strategies should be developed for use in areas where available data are less complete than in the RAPS area.

(4) Similarly, the development of a methodology for assessing the resultant costs in terms of lost production and increased production costs of proposed strategies will be appropriate.

(5) Investigations on the basis of study and experience in the St. Louis region should be carried forward to identify the interaction of local government and other instutional aspects that are relevant to the formulation of effective control strategies and their enforcement.

In general the detailed tasks, covered within the 405 series are scheduled well along in the Research Plan at a relatively low level of effort. Accordingly, their ultimate detailed content tends to be somewhat more speculative than tasks presented elsewhere in the Research Plan so that presentation of their content in detail does not appear warranted. Broadly, however, the tasks included in the 405 series are as follows:

405.1 Liaison and
Interaction with other Environmental Control
Programs

405.2 Techniques of Assessing
Social and Economic Factors

405.3 Methodology of
Assessing Operational Costs of
Control Strategies

405.4 Methodology of
Assessing Resultant Costs of Control Strategies

405.5 Institutional Aspects